Single quantum dots as local probes of electronic properties of semiconductors

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We have investigated size- and excitation-intensity-dependent changes of the luminescence spectra of single $In_{0.14}Ga_{0.86}As/GaAs$ quantum dots with diameters between 200 and 40 nm. The dot sizes investigated range from effectively two-dimensional structures down to diameters which are comparable to the length scales of electronic excitations, e.g., the excitonic Bohr radius, or of compositional fluctuations at interfaces. For dots occupied by single excitons, the luminescence linewidth decreases continuously with decreasing diameter. Using model calculations we relate this behavior to compositional fluctuations and obtain an estimate for their characteristic length scale. If the dots are occupied by several excitons the luminescence linewidth increases systematically with decreasing size. We describe this behavior by a model of carrier-carrier interaction effects on the luminescence linewidths. [S0163-1829(96)12627-8]

Optical studies of semiconductor quantum dots have received considerable attention recently.[1–7] In these structures all degrees of freedom of electrons and holes can be suppressed by a three-dimensional confinement potential. This results in the replacement of the quasicontinuous dispersion relations of electrons and holes in bulk semiconductors by sets of discrete levels similar to the eigenstates of atoms.

There are important differences between the electronic states and the optical transitions in an atom and those in a semiconductor quantum dot. For example, in a hydrogen atom there is a strong unscreened interaction between the nucleus and the electron. The Coulomb interaction between an electron and a hole in a semiconductor, on the other hand, is screened by the valence electrons of the atoms forming the solid. The high dielectric constants and the low effective masses lead to the formation of Wannier excitons that extend over many elementary cells of the semiconductor crystal and experience spatially varying properties on this scale.

The linewidths of optical transitions give information about fundamental properties of solids including the effects of potential fluctuations. In previous work involving optical studies of three- and two-dimensional systems, the areas probed generally are much larger than the potential fluctuations themselves. The averaging over many fluctuations results in an inhomogeneous broadening of the linewidths. In most of these studies it is therefore not possible to obtain microscopic information about the locally varying potentials associated with individual fluctuations.

In the present work we have studied optical transitions in single quantum dots with systematically varying sizes down to the order of the exciton diameter. We demonstrate for the first time that these single quantum dots are powerful probes of semiconductor properties on these microscopic length scales. Lateral quantization in a quantum dot implies that the carrier wavefunction extends effectively over the entire structure. This permits us to use the luminescence of the excitons as a monitor for semiconductor properties on a sub-100 nm length scale. In particular, the linewidth of the emission is sensitive to compositional fluctuations or to fluctuations in the spatial distribution of charges in the dot. By analyzing the experimentally observed changes of the linewidths versus the dot diameter with microscopic models for the cases of low and high excitation, we identify the dominant broadening mechanisms and obtain values for the relevant lengths scales.

The free-standing single quantum dots were defined by low-voltage electron-beam lithography. For luminescence studies of individual quantum dots the distance between adjacent dots was selected to be 20 μm (for microluminescence experiments at 35 K) and 50 μn (for 2-K luminescence experiments in a pumped He cryostat). The dots were wet etched into a 5-nm-wide $In_{0.14}Ga_{0.86}As/GaAs$ single quantum well.[8] For the In concentration and the quantum-well thickness used here, there is only one bound state in the conduction band of the quantum well. Scanning electron microscopy (SEM) micrographs of the dots indicate that they have a cylindrical shape. Thus, the energy states involved for optical transitions in the dots are described by radial and orbital angular momentum quantum numbers, n_r and m, respectively, in addition to the quantum-well subband index n_z.[7]

The single dots were excited by a cw Ar^+ laser with excitation densities between 0.1 and 10 W/cm^2 focused to about 5 and 20 μm for microluminescence and low-temperature luminescence experiments, respectively. The emission was dispersed by a 0.3-m monochromator and detected by a liquid-nitrogen-cooled charge-coupled device camera. The diameters of the dots were measured using high-resolution SEM micrographs with an accuracy of about 3

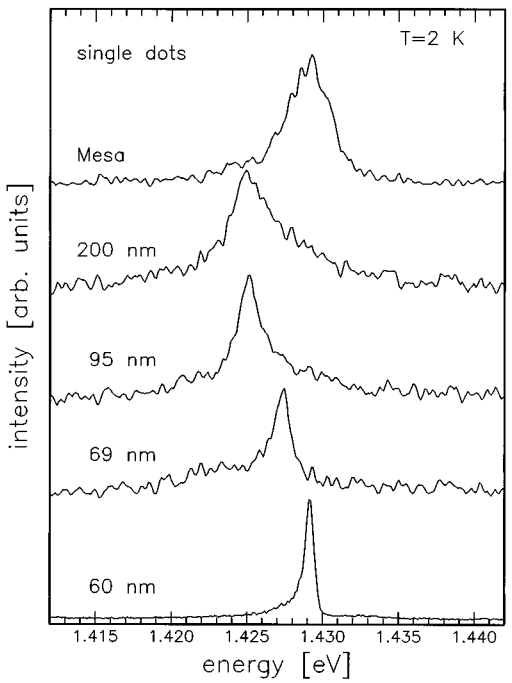

FIG. 1. Luminescence spectra of free-standing single quantum dots of varying diameters at low-excitation power in comparison to data taken from a large two-dimensional mesa structure.

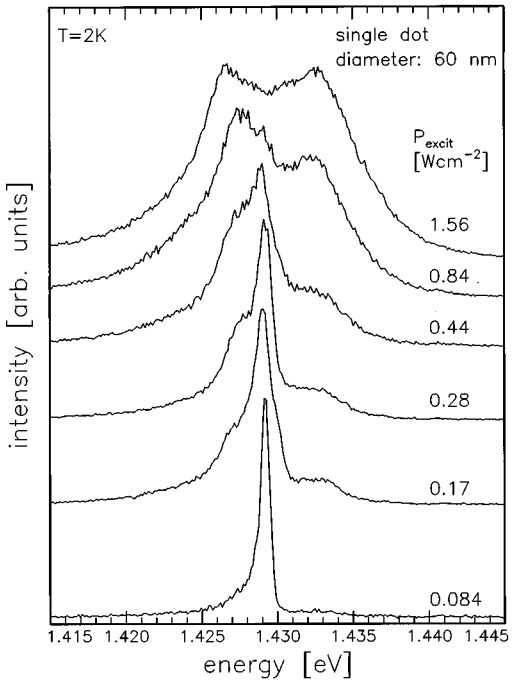

FIG. 2. Emission spectra of a 60-nm-diam dot for varying excitation powers ($T=2$ K).

nm. The spectra have been integrated for times between 1 and 30 s; i.e., all spectra are due to a large number of recombination processes in the dots.

Figure 1 shows low-excitation spectra of single quantum dots with varying diameters in comparison with spectra from a two-dimensional quantum-well reference, all taken at a temperature of 2 K. The redshift between the emission of the reference mesa and that of the 200-nm-diam dot is due to the relaxation of the biaxial strain in the etched dots. When the dot size is reduced below about 100 nm, we observe a shift of the emission line to higher energy due to the lateral quantization. This shift can be described well by calculations assuming a cylindrically symmetric lateral confinement.

Simultaneously with the increase of lateral quantization effects, we observe a continuous reduction of the luminescence halfwidths with decreasing dot size. For the 200-nm-wide dot the emission line halfwidth is about 3 meV and is comparable to the width of the two-dimensional reference. The 95-nm-diam dot has a significantly smaller halfwidth of about 2 meV. For the 60-nm-wide dot the halfwidth is reduced to about 0.75 meV, i.e., to about one-quarter of the spectral width of the reference emission.

Figure 2 shows a set of luminescence spectra from a free-standing 60-nm-diam dot for increasing excitation intensities. At the smallest excitation density of 0.084 W/cm^2 the luminescence consists of a single narrow line. Within the area of a dot of about 10^{-10} cm^2, 10^8–10^9 carrier pairs are generated per second. Using a typical exciton lifetime of about 100 ps, this implies that the dot is occupied for about 1–10 % of the time by one electron-hole pair. For the remaining time, the dot is unoccupied. We therefore associate the narrow line which dominates the low-intensity spectra with the recombination of single excitons.

With increasing excitation power a shoulder appears on the low-energy side followed at higher excitation by a feature on the high-energy side of the emission, indicating the simultaneous occupation of the dot by several electron-hole pairs. The lowest quantized dot level is characterized by the quantum numbers $n_z=1$, $n_r=1$, and $m=0$. Including the spin, the ground state of the dots may be occupied by up to two excitons which can form a biexciton. Due to the attractive interaction between the two excitons, the corresponding emission occurs at lower energy than the single exciton transition.[9,10] From the difference in the emission energy of the single exciton line and the low-energy transition of an electron-hole pair within a biexciton (shown, e.g., in the second and third spectrum from the bottom of Fig. 2), we determine a biexciton binding energy of about 2 meV for this dot size. For dots with larger diameters we observe a systematic decrease of the biexciton binding energy down to about 0.8 meV, which agrees with typical values for III-V quantum wells.[12]

If more than two electron-hole pairs occupy the quantum dot simultaneously, the Pauli principle requires the filling of higher quantized levels. The first excited dot level has quantum numbers $n_z=1$, $n_r=0$, and $m=\pm 1$ and can be occupied by up to four electron-hole pairs. For the 60-nm-diam dot the emission from the first excited state is located about 4 meV higher in energy than the emission from the ground state (see the upper traces in Fig. 2). It is interesting to note that the halfwidths of the transitions at high excitation powers are significantly wider than the emission lines of the single excitons observed at low excitation.

Most remarkably, the emission linewidths at high excitation powers show the opposite dependence on the dot diameter than those observed at low excitation. Figure 3 displays emission spectra of single $In_{0.14}Ga_{0.86}As$ dots of different

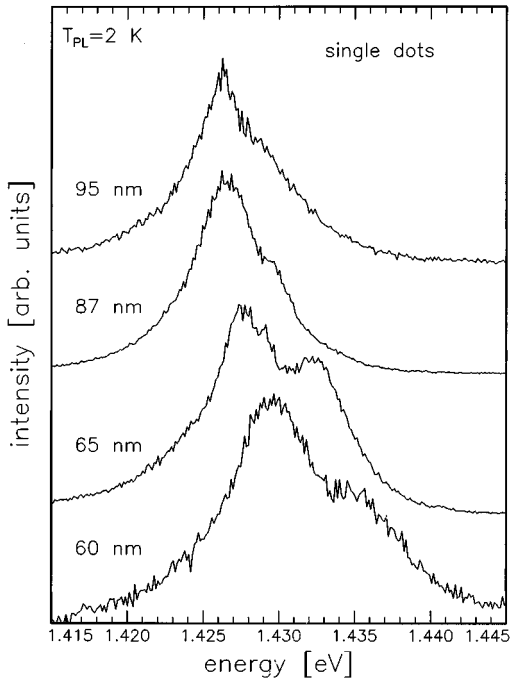

FIG. 3. Luminescence spectra of single quantum dots of varying diameters for high-excitation powers (about 2 W cm^{-2}). As a result of the high excitation, the dot ground state is filled with two electron-hole pairs and the first excited state is partially filled.

diameters recorded for excitation powers which result in a complete filling of the ground state and a partial occupation of the first excited state (about three electron-hole pairs per dot at all times). As can be seen from this figure, the width of the emission line due to recombination from electron-hole pairs in the ground state as well as the width of the feature at higher energy due to recombination of carriers in the first excited state increase continuously with decreasing dot size.

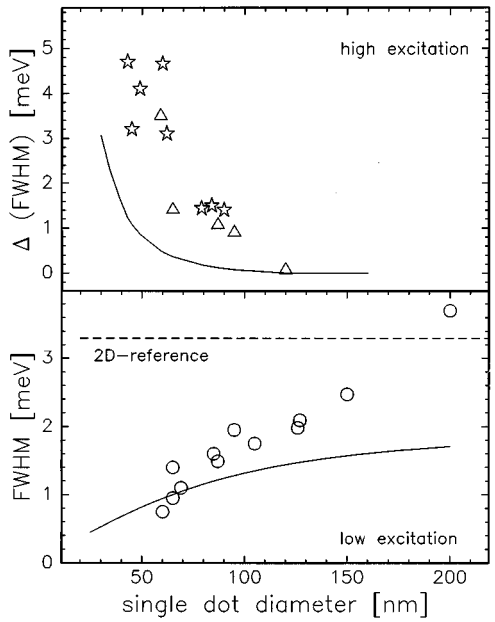

FIG. 4. Dot diameter dependence of the full width at half maximum of the luminescence linewidths at low excitation (about 0.1 W cm^{-2}, bottom) and at high excitation (about 2 W cm^{-2}, top). Circles and triangles: experimental data recorded at 2 K, stars: data taken at 35 K.

Figure 4 summarizes the experimental data for the dot diameter dependences of the emission linewidths at low and high excitation intensity (different symbols). In the low-intensity case the emission is due to individual excitons occupying the quantum dots. The high-intensity data give the emission linewidths under the experimental conditions of Fig. 3. Here the halfwidths of the transitions were obtained by fits of the emission spectra with Lorenzians. At low-excitation intensity we observe a reduction of the halfwidths of the luminescence line by a factor of about 4 as the dot diameter is reduced from 200 to 60 nm. For high-excitation intensity, on the other hand, Fig. 4 shows an increase of the halfwidths by a factor of about 2 as the dot diameter is decreased from 120 to about 40 nm.

The dot size dependences of the linewidths in the cases of both low and high excitation can be used to understand the underlying broadening mechanisms. At low excitation, emission linewidths in quantum-well systems are often associated with spatial variations in the widths of the quantum wells. In addition, in the case of an alloy there can be contributions to the linewidths from alloy scattering. In a dot structure, the contributions of these scattering mechanisms will decrease if the dot diameter becomes comparable to the length scale of the fluctuations. We have calculated the linewidths of the quantum dots that arise from monolayer fluctuations of the well width. Elastic or quasielastic scattering between the closely spaced dot states due to static potential fluctuations will give rise to dephasing of these states and to contributions to the homogeneous linewidths. The dephasing linewidth of a free quantum-well exciton with momentum k and center of mass energy $E_x=\hbar^2k^2/2M_x$ due to scattering from static cylindrical potential fluctuations of diameter d and magnitude δV is given by[11]

$$\Gamma_k=\left[\frac{2\pi M_x d^2(\delta V)^2}{\hbar^3\sigma}\right]\int_0^{\pi/2}\left[\frac{J_1(kd\cos\theta)}{2k\cos\theta}\right]^2 d\theta. \qquad (1)$$

Here M_x is the effective mass of the exciton, J_1 is the first-order Bessel function, and σ^{-1} is the number of such scattering centers per unit area.

Transmission electron microscopy (TEM) and growth data[13,14] suggest that a characteristic length scale of well width and compositional fluctuations at many III-V interfaces is in the range of 10–30 nm. We obtain the magnitude of the potential $\delta V=3.7$ meV from the variation of the ground-state energies of the electron and hole due to variation of the quantum-well width by 1 M. The center-of-mass exciton energy E_x is determined by the lateral quantization of its motion in the quantum dots. The dot size dependence of the linewidth is thereby related to the dependence of the scattering probability on the quantized energy of the dot and to the characteristic size of the fluctuations.

The solid line in the lower panel of Fig. 4 has been calculated within the above model for a characteristic diameter of the fluctuations of 20 nm and an areal density of scattering centers of 2.3×10^{10} cm^{-2}. The dot size dependence as well as the overall magnitude of the measured emission line-

widths are given well by this model using parameters that are physically reasonable and that are consistent with other data.[13,14]

In order to understand the increase of the emission linewidths with decreasing dot size at higher excitations, we have calculated the elastic dephasing scattering rates of carriers in the quantized dot states due to Coulomb scattering from other occupied states of the dot. For the present purposes we treat the occupied states as static charges. We calculate the confined states in the effective-mass approximation. Between $In_{0.14}Ga_{0.86}As$ and GaAs a conduction-band discontinuity of 97 meV and a valence-band (heavy-hole) discontinuity of 60 meV was used, and the potential offset at the etched vacuum surfaces was taken to be infinite. The resulting electrostatic potential arises from the different extensions of the electron and hole wave functions into the GaAs barriers. For two electron-hole pairs in the lowest radial state the potential is

$$\Phi(\vec{r}) = 2\frac{e^2}{4\pi\varepsilon\varepsilon_0}\frac{1}{[aJ_1(x_{01})\sqrt{\pi}]^2} \times \int_{-\infty}^{\infty} dz' \int_0^{2\pi} d\phi' \int_0^a \rho' d\rho' \frac{J_0^2\left(\frac{x_{01}\rho'}{a}\right)\zeta^2(z')}{|\vec{r}-\vec{r}'|}. \quad (2)$$

Here a is the radius of the cylindrical dot, J_0 is the Bessel function of zeroth order, x_{01} is its first zero, and $\zeta^2(z)$ gives the z dependence of the net charge density of the occupied states. The potential is then calculated by direct numerical integration of Eq. (2).

The solid line in the upper part of Fig. 4 shows the contribution of carrier-carrier scattering to the linewidth as calculated from Eq. (2) for a process in which an electron and a hole each scatter from their $m=+1$ states to their $m=-1$ states due to interactions with static charges, which were taken to be two electron-hole pairs in the dot ground states. It should be noted that for any number of occupied states the contribution to the linewidth increases with decreasing dot size. This dependence arises from the $(1/a^2)$ factor in Eq. (2), which comes from the normalization of the quantized wave function in the quantum dot. The comparison of experimental data and calculations displayed in Fig. 4 shows good agreement regarding the size dependence as well as the overall magnitude. A more complete analysis of the magnitude of the linewidths under high excitation would require averaging the different carrier occupations of the dot that occur in the experiments and including the effects of disorder.

In summary, we have used semiconductor single quantum dots occupied by a few optically excited electron-hole pairs as local probes for compositional fluctuations and carrier-carrier interaction effects. We have related the experimentally observed dot size dependence of the linewidths to specific physical mechanisms in both the regimes of low and higher excitation. Our studies show that single nanostructures may serve as powerful microscopic probes for investigations of a wide variety of physical phenomena, including the coherence of electronic and vibronic excitations, exciton-exciton, and carrier-carrier interactions on the length scales of disorder effects in a wide variety of material systems.

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