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Effects of few-particle interaction on the atomiclike levels of a single strain-induced quantum dot

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We investigate the effects of few-particle population of a single strain-induced quantum dot by optical excitation. The low-power photoluminescence spectra consist of sharp lines with energy separation of a few meV, associated to the formation of excitonic molecules in the single dot. With increasing photoexcitation intensity, the population of higher states is observed; however, we also observe a clear intensity dependence of the transition energies, inconsistent with a simple filling of exciton levels. Based on a theoretical model for interacting electron-hole pairs in the dot, we obtain good agreement with experiment and show that exciton-exciton interactions control the spectral changes as the number of pairs is increased.

The realization of high-quality semiconductor quantum dots has recently lead to the demonstration of artificial atoms, in which addition energies[1] and Zeeman splitting[2] associated with three-dimensionally confined electron states mimic the characteristics of natural atoms. Similarly, the optical excitation of a single quantum dot (QD) results in optical spectra reminiscent of atomic transitions with sharp and spectrally narrow lines.[3–7] The understanding of the complex optical spectra from single dots and their dependence on photoexcitation intensity is, however, still quite poor. The atomiclike lines in the spectra were found to exhibit intriguing red- and blue-shifts, and new features are seen to appear and disappear depending on the number of injected electron-hole pairs.[8–10] Such a complicated dynamics originates from the Coulomb correlation among pairs of carriers (excitons) confined in the dot, which is often neglected in the analysis of these experiments. In fact, we expect that whenever an electron-hole (e-h) pair is added to the dot, the energy states must change because of the resulting additional Coulomb interactions.

In this paper, we report a complete study of the single-dot spectra, both experimental and theoretical, which elucidates the atomiclike nature of the optical transitions and the important role of Coulomb correlations in an isolated quantum dot. As a prototype nanostructurewe have chosen a single strain-induced quantum dot physically isolated from the surrounding environment in a nanomesa defined by high-resolution lithography. This system has never been investigated by local spectroscopy so far, but for an intriguing near-field luminescence experiment in which no atomiclike features were observed,[11] despite the excellent zero-dimensional characteristics shown recently by the same samples.[2,12–14] The photoluminescence (PL) of the single dot is measured with increasing the number of e-h pairs, ranging from 1 to approximately 20. At low injection powers, corresponding to an average number of only a few e-h pairs, we do observe the appearance of correlated exciton lines, associated with the formation of biexcitons, on the low-energy side of the ground level transition. With increasing particle number, the energy and the relative intensity of these transitions are modified due to multiexciton interaction processes, and state filling of the higher quantized levels occurs. A theoretical model of the luminescence process is developed to interpret these data. Within a direct-diagonalization scheme,[20] we compute the many-particle states for an increasing number of interacting e-h pairs. The calculated luminescence spectra are in very good agreement with the experiment.

The single dot structure was fabricated by atomic-force microscopy (AFM) and nanolithography on a QD sample grown by metal-organic vapor phase epitaxy (MOVPE). The sample consists of self-organized InP islands (stressors) grown on an 8 nm thick $In_{0.08}Ga_{0.92}As/GaAs$ quantum well (QW) (the barrier thickness is 20 nm). The InP islands induce a paraboliclike strain potential that laterally confines the carriers in the InGaAs QW underneath the stressor. The typical size of the InP stressors is of the order of 80 nm in diameter and 25 nm in height, with a measured areal density of 10^9 cm^{-2}. The resulting ground state wave function in the QD's exhibits typical lateral extension of about 30 nm (at 1/e of its maximum amplitude), and extends over approximatively 50 nm, which is the lateral size of the strain-induced parabolic-like potential.[12] Details on the samples can be found in Refs. 13 and 14. To isolate a single dot, a square window of 10×10 μm^2 was first opened into a gold mask deposited on the sample surface by UV lithography and lift-off. Then a 200×200 nm^2 mesa, containing a single quantum dot, was defined by AFM lithography and HCl wet chemical etching at the center of the 10×10 μm^2 window. The spectra of the single dot were excited by an argon laser

and detected by a charge-coupled device (CCD) mounted on a 0.32 m spectrometer. In order to probe the effects of sample heating, different measurements were performed by reducing the average laser power by means of a mechanical chopper. No difference in the microphotoluminescence (micro-PL) spectra recorded with different duy cycles were observed for exciting power densities between a few μW and 40 μW. The measurements were performed in a modified vibration-free closed cycle He cryostat where the cold finger was mechanically decoupled by the last He pumping stage through a copper ribbon. The vibrations at the sample were measured by an interferometric method and amounted to ± 0.1 μm, i.e., well within the spot size ($\simeq 0.6$ μm). A flat cryostat head was built to allow backscattering PL experiments with a short focal distance objective. The minimum temperature reached by such a system was 25 K. A confocal micro-PL setup was used to increase the ratio between the signal and the background noise level. The spectral resolution of the experiments was of the order of 0.7 meV. This experimental technique has several advantages over other methods reported in the literature. First of all, the strain-induced quantum dot is physically isolated from the surrounding dots (which were completely removed). This is not the case in confocal PL experiments[6,8–10] or scanning near-field optical microscopy (SNOM) experiments[7,11] carried out on two-dimensional arrays of dots, where carriers can diffuse and eventually populate neighboring dots.[15] Second, the dot forms in the quantum well under the InP stressor, far from the sample surface and from the sidewalls, so that no surface states and depletion layers affect the recombination. This is not the case in dots that are isolated through a metallic mask,[16] where the Schottky barrier may change the band alignment, or in free-standing dots that are affected by charges at the surface.[17]

In Fig. 1(a) we show the space-integrated PL spectra of the original unprocessed sample, containing a large population of dots, as a function of the excitation intensity. The PL spectra consist of four broad lines associated to recombination from the ground state (1Σ) and the first three excited states (1Π, 2Σ, 2Π, respectively) of the dots. The band centered at 1.464 eV is due to the QW connecting adjacent dots. With increasing excitation power, the relative intensity of the four lines changes, but their energy position and full width at half maximum (about 7 meV) remain fixed. The intensity dependent micro-PL spectra of the single dot isolated in the mesa is shown in Fig. 1(b), in the energy range of the dot emission. At low photoexcitation power, the full width at half maximum of the observed lines is now reduced to 0.8 meV. Even though such a linewidth is comparable to the spectral resolution of the experiment and is limited by the sample temperature (about 25 K), we believe that the relatively large linewidth is typical of single QD structures fabricated starting from an InGaAs quantum well,[17] where photoexcited surface charges or near surface impurity states can generate fluctuation of the QD potential in time.[17,11] Moreover microscopic interface defects and In fluctuation in the original QW can contribute to the single QD line broadening.[18] In our case, the ground level wave function probes small compositional fluctuations in the ternary alloy well underneath the InP dot, where surface charges can eventually accumulate and affect the dots buried under the stressor. In a single-particle picture, we can say that at the lowest

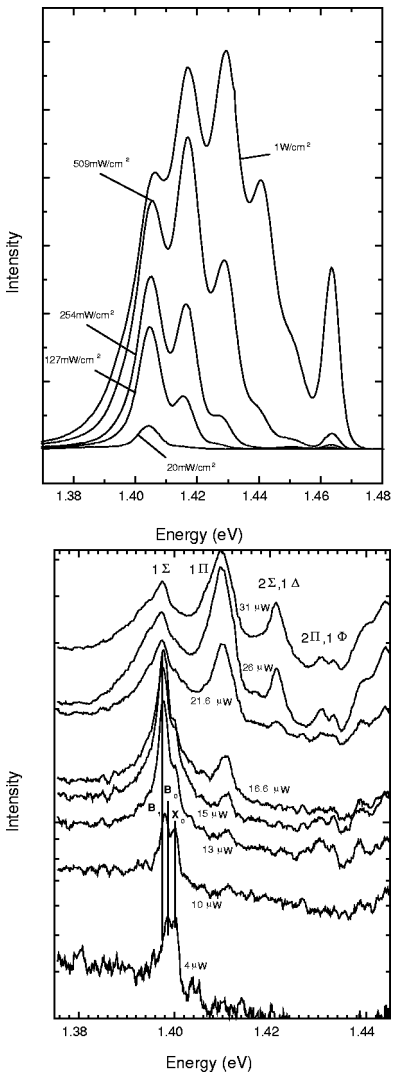

FIG. 1. (a) Space integrated photoluminescence of the original unprocessed sample containing a large population of quantum dots. Labels indicate the photoexcitation power for each curve. (b) Microluminescence spectra of the sample with a single quantum dot isolated in a mesa, plotted for different values of the photoexcitation power. Only the lowest energy range is shown, associated to emission from the dot.

density only the ground state 1Σ is populated. This state has twofold degeneracy, and therefore can host up to two electrons and two holes with antiparallel spins. The sharp lines X_0 and B_0, that are split by about 1.5 meV, can then be tentatively associated to the single-exciton and the biexciton level resulting from their interaction. A thorough discussion of this assignment will be given later. With increasing excitation density the B_0 line becomes dominant, red shifts and broadens, probably overwhelming an additional low-energy line (B_1 see below). In any case, with increasing the number of excitons in the dot, its emission intensity raises superlinearly. When the 1Σ-related emission lines saturate, the emission from the first excited state increases. Also in the 1Π emission we note an asymmetry in the low energy tail of the line, due to the contribution of different multiexciton lines. In this case we cannot resolve the different contributions due to the higher number of interacting particles that populate the first excited state (four electrons and four holes). A similar

behavior is observed when the second and third excited states start to be populated with increasing photoexcitation density. Note that the peaks are strongly asymmetric, indicating that they originate from the convolution of several lines, as opposed to the perfect Gaussian shape of the peaks in the space-integrated PL [Fig. 1(a)], associated with inhomogeneous broadening due to the dot size distribution. In the following, we focus on the few-exciton regime [$P \leqslant 20$ μW in Fig. 1(b)] and investigate the unexplained structure and density dependence of the lines associated to the lowest state.[19] To understand the origin of the observed lines, we introduce a description of the luminescence process that takes into account the interaction between e-h pairs. From a theoretical point of view, luminescence involves a process where one e-h pair is removed from the interacting many-particle system, and one photon is created. Thus, the luminescence spectra provide information about e-h excitations of the interacting many-particle system. More specifically, the luminescence spectrum $I(\omega)$ can be computed as[20]

$$I(\omega) \propto \sum_{n,n'} f_T(E_n^N) |\langle n';N-1|\hat{\boldsymbol{\pi}}|n;N\rangle|^2 \mathcal{D}_\gamma(\omega - E_n^N + E_{n'}^{N-1}). \tag{1}$$

Here $|n;N\rangle$ is the nth excited state of the interacting electron-hole system with energy E_n^N (N being the number of e-h pairs), $\mathcal{D}_\gamma(\omega) = 2\gamma/(\omega^2+\gamma^2)$ with the phenomenological damping constant γ accounting for interactions with the QD environment (e.g., phonons or fluctuations of the electric fields), and $\hat{\boldsymbol{\pi}}$ is the interband polarization operator describing the light-matter coupling within the usual dipole and rotating-wave approximations. In Eq. (1) we have assumed that before photon emission the interacting electron-hole states are occupied thermally according to Boltzmann's distribution at temperature T, $f_T(E)$. In our calculations for the QD, we start from the single-particle states derived by solving the single-particle Schrödinger equation within the envelope-function and effective-mass approximations, by means of a plane-wave expansion with periodic boundary conditions.[21,22] For electrons and holes, respectively, we keep the 10 energetically lowest single-particle states, and restrict the many-particle basis to the $\approx$100 Slater determinants of lowest single-particle energies. Within these bases, we obtain the many-particle states by direct diagonalization of the Hamiltonian matrix; details of our computational approach will be published elsewhere.[23] Our theoretical approach is similar to earlier studies[24–26] where, however, only an extremely limited number of basis functions was considered. Figure 2 shows the luminescence spectra for an increasing number of e-h pairs, N, as computed from Eq. (1). For a single electron-hole pair confined in the QD [Fig. 2(a)], the luminescence originates from the decay of the ground state exciton (X_0). For two electron-hole pairs [Fig. 2(b)] we observe the appearance of the biexcitonic line, which is shifted by $\sim$1.5 meV to lower energies (B_0); because of the finite temperature ($T=25$ K) considered in our calculations, an additional small peak appears at lower photon energy attributed to the decay of a biexcitonic resonance (B_1). For even higher exciton concentrations [Figs. 2(c–e)] we observe luminescence from both the 1Σ and 1Π shells. Because of the finite temperature, we oberve an asymmetric broadening of

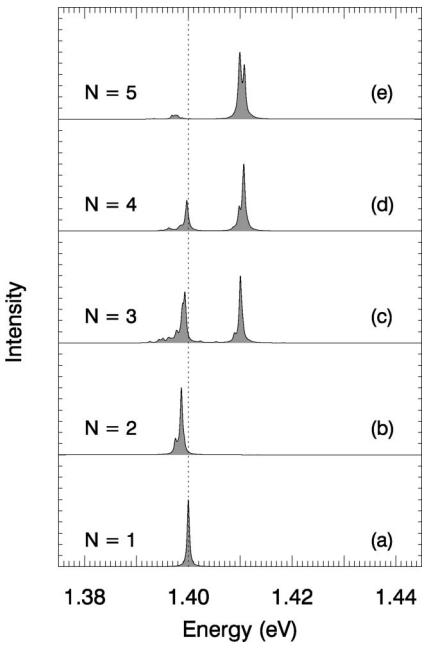

FIG. 2. Luminescence spectra for an increasing number of excitons, N, as computed from Eq. (1); we use $T=25$ K and $\gamma = 0.25$ meV. Photon energy zero has been chosen according to the energy of the ground state exciton. In our calculations we assume, for simplicity, that the interacting electron-hole states consist of optically allowed excitons, X (i.e., excitons where electrons and holes have antiparallel spin orientations). More specifically, we assume: (a) $1X$; (b) $1X_\uparrow$, $1X_\downarrow$; (c) $2X_\uparrow$, $1X_\downarrow$; (d) mixture between $2X_\uparrow$, $2X_\downarrow$, and $3X_\uparrow$, $1X_\downarrow$; (e) $3X_\uparrow$, $2X_\downarrow$.

the emission peaks due to the convolution of several lines. It is worth noting that our theoretical results are in good agreement with those of a recent theoretical study of Hawrylak[25] where, however, only a rather simplified model system was considered. In stationary condition experiments, one is averaging over an ensemble of configurations with different numbers of excitons. As a function of increasing photoexcitation density, this ensemble average involves increasing contributions from spectra with higher number of excitons, N. Therefore, the evolution with power in the experimental spectra below 20 μW [Fig. 1(b)] can be interpreted in terms of increasing contributions from the spectra shown in Figs. 2(b) to 2(e). It is worth noting that both the calculated binding energy of the biexciton, as well as the asymmetric broadening due to the convolution of the several lines on the low-energy side of the B_0 resonance are in good agreement with the experimental findings. Moreover, the biexciton binding energy found here (1.5 meV) is consistent with the calculations reported in Ref. 17 for QDs fabricated by lithography and etching starting from a $In_{0.14}Ga_{0.86}As/GaAs$ quantum well. In our case the lateral geometrical extention of the QD is similar but the starting quantum well has an In content of 0.08 and a width of 8 nm resulting in a sligthly smaller exciton and biexciton binding energies. The good agreement between experiment and theory confirms that photoexcited electrons and holes are strongly confined in the quantum dot, which behaves as an artificial atom. We have shown that confinement induces not only a strong exciton binding, but also a significant coupling between excitons, that is essential

to describe the spectral changes as the number of e-h pairs is increased. We expect that this effect will influence the physics of QD-based optical quantum devices in general, and that it may be used to control the appearance and suppression of narrow spectral lines in these structures.

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