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Optical gain saturation effects in InAs/GaAs self-assembled quantum dots

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An attempt has been made to understand electronic structure and optical (lasing) properties of self-assembled InAs/GaAs quantum dots (QD) and to describe saturation effects in QD levels population. The new, improved rate equation model has been developed. The impact of carrier relaxation and level depopulation inside quantum dots on lasing properties, in particular on gain depressing, is discussed.

1. Introduction

The carrier recombination processes in self-assembled InAs/GaAs quantum dots (QDs) have recently been a subject of an intensive investigation because of their important applications in optoelectronic devices such as light emitting diodes and lasers [1]. However, the size fluctuations and the shape nonuniformity of the QDs result in gain spectra which are much broader than those usually observed in quantum-well (QW) active media [2]. Additionally, due to the limited number of states available in quantum dots, gain saturation effects are inevitable in quantum dot lasers. In particular, in QD vertical cavity surface emitting lasers (VCSELs), a higher gain is needed because of shorter cavities, comparing to edge emitting lasers, and there the laser action tends to occur from excited states at higher photon energies. Thus the evolution of the spectral distribution of emission intensity and of optical gain with excitation (current or optical pumping) is of importance for designing QD lasers.

2. Experimental results

In this paper we present results of the spectral distribution of spontaneous emission intensity study of self-assembled InAs/GaAs quantum dots for GaAs based emitters at 1300 nm. The self-assembled QDs have been grown by molecular beam epitaxy by depositing 8.9 Å of InAs on top of GaAs and capping with a 5 nm-thick $In_{0.15}Ga_{0.85}As$ layer. The details concerning growth procedure and structural characteristics of QD structures studied can be found in [3]–[5]. The plane view transmission electron microscope (TEM) picture of QD structure is shown in Fig. 1.

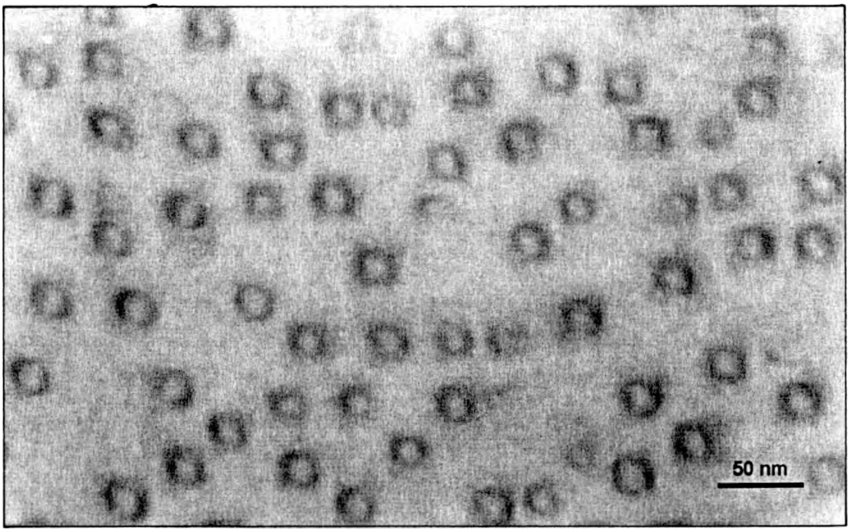

Fig. 1. Plane-view TEM image in [001] direction of InGaAs QDs embedded in GaAs matrix. The diameter of QDs is approximately 20–30 nm and their surface density is 4.5×10^{10} cm^{-2}.

The photoluminescence (PL) spectra of the sample with QD surface density equal to 4.5×10^{10} cm^{-2} for different excitation intensities, at 77 K and 300 K are shown in Fig. 2 and Fig. 3, respectively. Three peaks from the QDs are clearly resolved at both temperatures. They refer to the ground state and to the first two excited electron-hole states. A peak at around 870 nm at 300 K, observed at high excitation intensities is related to GaAs. Also, a weak ~990 nm emission from the QW formed by $In_{0.15}Ga_{0.85}As$ wetting layer can be seen at 300 K spectra. The emission peaks are almost equally spaced (60–62 meV), regardless of the measurement temperature, which suggests that in-plane (x, y) confining potential is harmonic.

3. Electronic structure of InAs/GaAs quantum dots

The detailed theoretical analysis of the electronic structure of QDs containing up to 10 electrons (holes), using the self-consistent Hartree method, has been

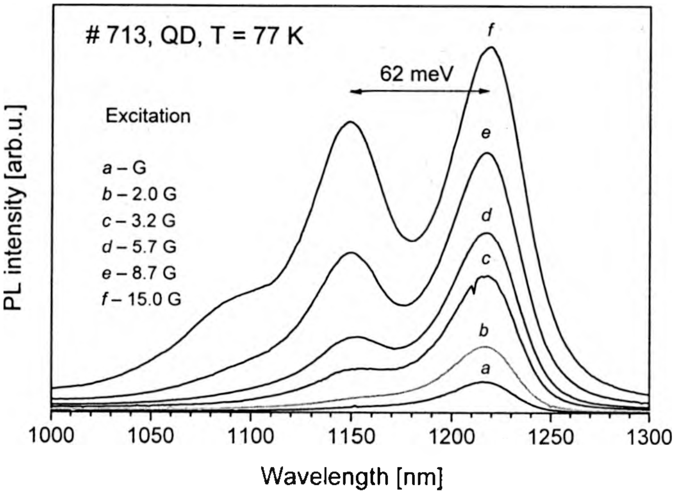

Fig. 2. Spontaneous emission *vs.* wavelength at 77 K for different excitation intensities (G – excitation density, $G = 10^5$ W/m^2).

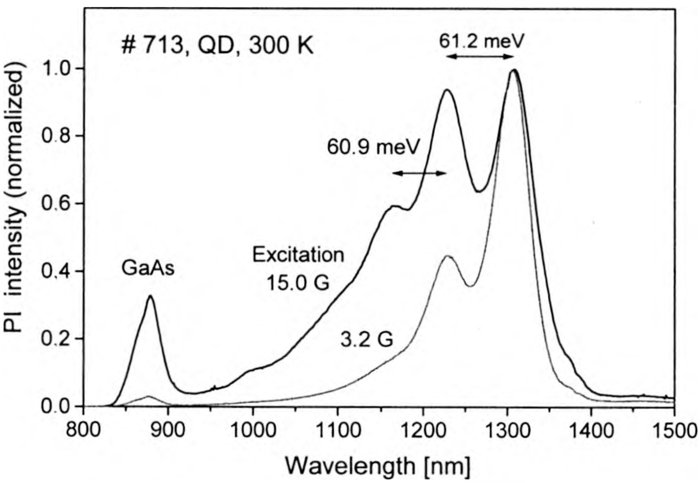

Fig. 3. Spontaneous emission *vs.* wavelength at 300 K for different excitation intensities.

performed. The bare dot potential has been represented by finite QW potential in the growth direction z and 2D harmonic potential in xy-plane. The idea of the Hartree method relies on interactive solving of the set of N effective mass equations for N electrons in a quantum dot. Each equation takes into account the effective potential energy which is unique for the electron being considered. This allows us to consider the influence of the electron charge on the quantum dot properties [6]. The charge of the confined electrons modifies in an essential way the energy spectrum of the dot, as well as the shape of the confining potential. A simplified Hartree method (assuming that all electrons in a quantum dot feel identical potential), which greatly speeds up calculations has been also tested. It is particularly useful when maximum material gain is to be derived for further use in laser simulations.

Calculated electron and hole levels for quantum dots with different occupations are shown in Figs. 4 and 5. The levels shift to higher energy with charging quantum dots. When dots are occupied by both electrons and holes, the level shift depends on the net charge which generally is small (majority of dots are electrically neutral).

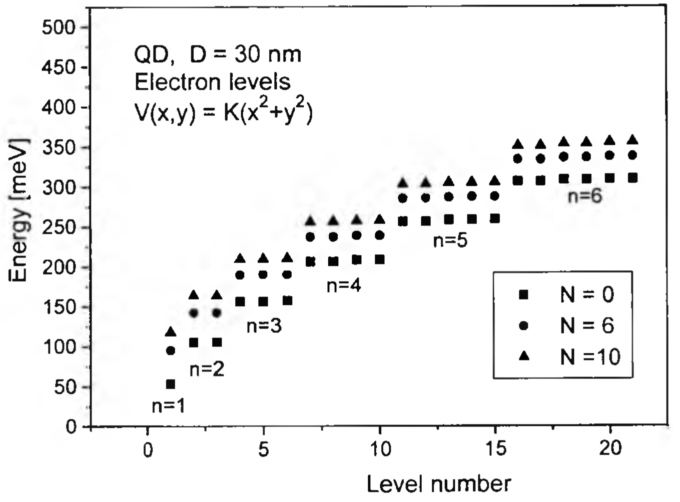

Fig. 4. Electron energy levels in InAs/GaAs QD calculated by the self-consistent Hartree method for different dot occupations (n – level number, N – number of electrons in QD). $V(x,y)$ – quantum dot harmonic potential, $K = 3\cdot 10^{14}$ V/m^2.

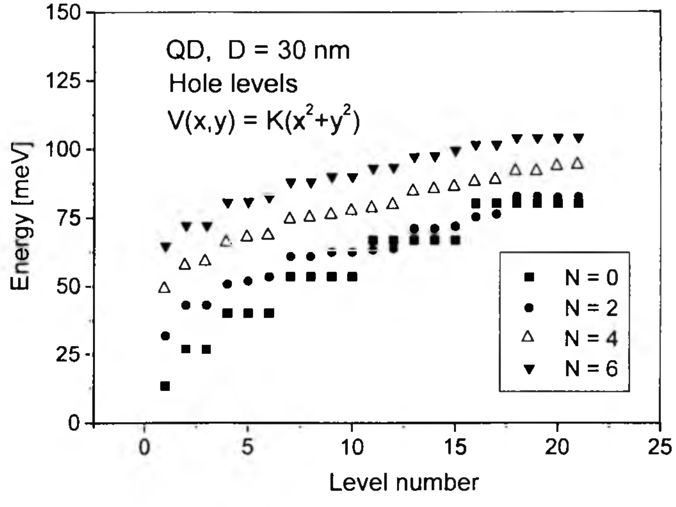

Fig. 5. Hole energy levels in InAs/GaAs QD calculated by the self-consistent Hartree method for different dot occupations (n – level number, N – number of electrons in QD).

So the results of calculations confirmed that despite the influence of the charge of electrons accumulated in QD on the energy levels, in the case when electrons and holes are simultaneously present, the electrostatic effects tend to cancel out and energy separation between electron–hole transitions referring to different principal quantum numbers remains almost the same (within a few percent). The geometrical parameters of QD, used in calculations, were taken from TEM measurements. The average quantum dots were 20–30 nm in diameter (in xy-plane) and 7–10 nm in hight. Due to the alloy intermixing effects the dots were not pure InAs, but rather varying in composition InGaAs in GaAs matrix. Therefore, the true extent of the in-plane harmonic potential was roughly twice the diameter. Since the confinement in z-direction is much stronger than in xy-plane, it does not influence the detailed structure of the levels and produces only a rigid shift of the whole energy spectrum (only $n_z = 1$ level is present in QD). Calculations showed that electronic structure resulting from the assumed dot parameters agreed with spectroscopic measurements. Assuming the Gaussian distribution of QD size fluctuations, one can calculate the spontaneous emission characteristics of an inhomogeneously broadened quantum dot system which can be directly compared with PL measurements shown in Figs. 2 and 3. Knowing transition energies, the gain spectrum of the dot assembly can be calculated assuming the Lorentzian lineshape of the gain of an individual dot and Gaussian broadening due to the size fluctuations.

The PL spectra of the studied system show that the material gain is strongly asymmetric due to the contribution of excited QD states already at comparatively low excitation. It has also been found that well before the intensity of the energetically lower ground state saturates, the higher states start to radiate. This is a clear evidence of finite relaxation time and appears to be in clear contradiction to the belief that in the case when the separation between the upper level and the ground state of the system (~ 60 meV) is sufficiently high, the lasing from the excited state before the full saturation of the lower level is precluded [2].

4. Rate equation model – saturation effects

Next we will develop a rate equation model, describing the population of QD levels for arbitrary excitation intensity. Conventional rate equation models, which rely on the mean field theories, do not describe properly the level population in a quantum dot ensemble and thus cannot predict correct gain values at high pumping levels. We have developed an alternative model based on the assumption of the non-thermal distribution of carriers. We assume that the energy spectrum of transitions in QD consists of two parts. For low energies the spectrum is discrete with constant distance between levels (2D harmonic oscillator model); for higher – continuous with bulk density of states. The probability of interaction between a pair of levels (either relaxation or excitation) in a unit of time is decreasing with the gap between interacting levels

$$p = \frac{1}{\tau_r} \exp\left(-\frac{|E_1 - E_2|}{kT}\right) \tag{1}$$

where τ_r is the relaxation time, E_1 and E_2 are the energies of interacting states, T is the temperature of the crystal. As the interaction may mean both transition upwards (excitation) and downwards (relaxation), we need to determine components $p_\uparrow$ and $p_\downarrow$ satisfying $p = p_\uparrow + p_\downarrow$. The relation between $p_\uparrow$ and $p_\downarrow$ is established by the assumption that states in a non-lasing dot are populated in the manner described by Fermi–Dirac distribution. That yields the following relation:

$$p_{1\to 2} = \frac{p}{1+\exp\left(\frac{E_2 - E_1}{kT}\right)}. \tag{2}$$

Let M and N be numbers of respectively electron and hole energy levels. We are going to build a system of $M+N+2$ equations, describing population of each of $N+M$ levels and two reservoirs (carriers from the continuous part of the spectrum). Let x_i ($i = 1,\ldots,M$) and y_j ($j = 1,\ldots,N$) be the populations of states in relevant bands

$$-\sum_{k=1}^{M} kx_i(1-x_k)p_{i\to k} + \sum_{k=1}^{M} kx_k(1-x_i)p_{k\to i} - c_i^\uparrow x_i + c_i^\downarrow(1-x_i) - r_i(x_i, y_i) = 0,$$

$$i = 1,\ldots,M,$$

$$-\sum_{l=1}^{N} ly_j(1-y_l)p_{j\to l} + \sum_{l=1}^{N} lx_l(1-y_j)p_{l\to j} - v_j^\uparrow y_j + v_j^\downarrow(1-y_j) - r_j(x_j, y_j) = 0,$$

$$j = 1,\ldots,N,$$

$$-\sum_{l=1}^{N} ly_j(1-y_l)p_{j\to l} + \sum_{l=1}^{N} lx_l(1-y_j)p_{l\to j} - v_j^\uparrow y_j + v_j^\downarrow(1-y_j) = 0,\ j = M+1,\ldots,N,$$

$$N = \sum_{k=1}^{N} 2kx_k + N_{cc},$$

$$N = \sum_{l=1}^{N} 2ky_l + N_{vc}. \tag{3}$$

In the first three equations the leading terms describe depopulating (the second one populating) of i-th level by relaxation or excitation to (from) another level in the discrete spectrum of QD. Function r is responsible for radiative recombination. It depends on many parameters, but amongst them on the occupation of interacting levels, which is the most important for the model considered. Since we assume that the radiative recombination is possible only between states with the same quantum number, only M first levels in the valence band can be depopulated in such a manner. The last two equations just count carriers in both bands. Factors k and l under sums are due to degeneracy of 2D-harmonic oscillator levels. Terms $v_j^\uparrow$, $v_j^\downarrow$, $c_i^\uparrow$, $c_i^\downarrow$ describe interactions between the reservoir and relevant level in the valence and conduction band. They are calculated assuming Eq. (2) and Fermi–Dirac distribution in the reservoirs and of course depend on N_{cc} or N_{vc}. In the last two equations of the set (3) N is the overall number of carriers, N_{cc} and N_{vc}, are the numbers of carriers in the conduction and valence band reservoirs. The external excitation fills the reservoirs (wetting layer, barriers) at a specified generation rate. From the reservoirs the carriers are captured into the dots. We consider each separate capture of electrons and holes. We further assume that the dots are uncoupled. With the above assumptions we obtain numerically the steady-state solution of the model, under the condition of stimulated emission, which is illustrated in Fig. 6. The plot shows the occupation of the QD levels at 300 K as

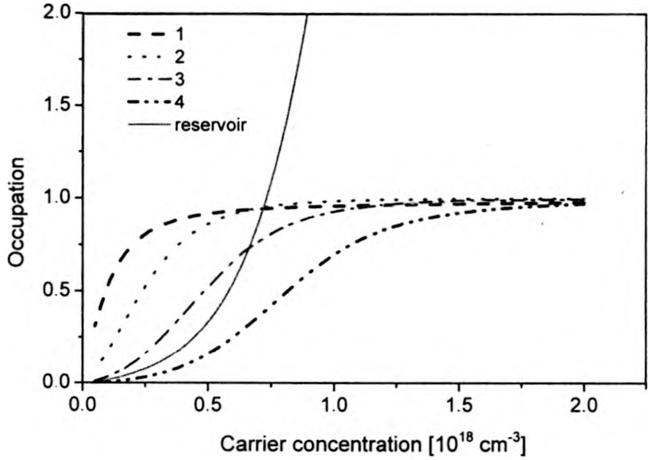

Fig. 6. Population of dot states E_n, $n = 1,\ldots,4$ as a function of external excitation density at 300 K.

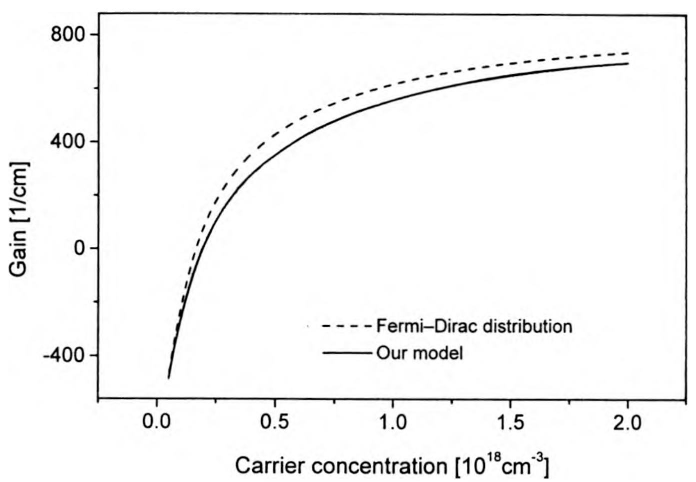

Fig. 7. Gain for the transitions between $n = 1$ levels in QD as a function of external excitation density at 300 K.

function of excitation density. We observe clear saturation of QD level population, although the occupation of higher laying levels starts to grow before the lower laying levels, including the ground state, saturate. This can have serious consequences for lasing and in particular can result in two-state lasing. Figure 7 shows the gain for transitions between ground states in QD as a function of carrier concentration, calculated under the assumption of the thermal (Fermi–Dirac) distribution of carriers in QD (dashed line) and calculated using our model (solid line). Due to depopulating of levels by stimulated emission, the gain in the realistic QD model is clamped down, comparing to the one for thermal distribution of carriers between QD levels. The specific magnitude of gain lowering depends on the ratio of stimulated lifetime to relaxation lifetime, assumed in the model.

5. Conclusions

We have studied electronic structure and optical properties of self-assembled InAs/GaAs quantum dots emitting at around 1300 nm (300 K). The structures containing dots were grown by molecular beam epitaxy. The detailed theoretical analysis of the electronic structure of quantum dots containing up to 10 electrons (holes), using the self-consistent Hartree method, has been performed and the results were compared with PL measurements. It has been found that bare dot potential can be represented by finite QW potential in the growth direction z and 2D harmonic potential in xy-plane. Numerical calculations have shown that despite the influence of the charge of electrons accumulated in QD on the energy levels, in the case when electrons and holes are simultaneously present, the electrostatic effects tend to cancel out and energy separation between electron–hole transitions referring to different principal quantum number remains almost the same. In an attempt to understand gain saturation in QDs we have developed the rate equation model, describing population of QD levels for arbitrary excitation intensity. We have observed clear saturation of QD level population, although the occupation of higher laying levels starts to grow before the lower laying levels, including ground state, saturate. This can have serious consequences for lasing and in particular can result in two-state lasing. Due to the depopulating of levels by stimulated emission, the gain in the realistic QD model is clamped down, comparing to the one for thermal distribution of carriers between QD levels.

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