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On the modulation mechanisms in photoreflectance of an ensemble of self-assembled InAs/GaAs quantum dots

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We present the investigation of the modulation mechanisms in photoreflectance (PR) spectroscopy of an ensemble of self-assembled semiconductor quantum dots (QDs). In order to distinguish between possible factors contributing to the total modulation efficiency of QD transitions, a photoreflectance excitation experiment has been performed on an InAs/GaAs quantum dot structure grown by solid-source molecular beam epitaxy. It has been observed that the intensity of PR features related to QDs changes in a function of the wavelength of the pumping laser, tuned from above-GaAs band gap down to below wetting layer ground state transition. Based on this dependence we have shown that most of the QD PR signal intensity originates from the modulation of the built-in electric field caused by carriers photogenerated in GaAs layers. We also conclude that the modulation of QD transitions related to a possible modification of the dot properties due to filling them with carriers is negligible in PR experiment on an ensemble of dots. An additional confirmation of the PR results has been obtained by using contactless electroreflectance (CER), demonstrating that the line shape of PR and CER QD resonances is almost identical in both spectra. Thus, the QD transitions can be analyzed by using the standard low field line shape functional form applicable in any electromodulation spectroscopy. © 2006 American Institute of Physics. [DOI: 10.1063/1.2355551]

I. INTRODUCTION

Semiconductor heterostructures containing layers of self-assembled quantum dots (QDs) have been a subject of thorough studies recently. The reason for this interest is twofold. First of all, QDs may be treated as a sort of laboratory to control and investigate many-body effects, resulting from the fact that a controlled number of carriers is confined in a small volume of semiconductor material.¹ Secondly, there are many device structures based on QDs that have improved parameters in comparison with other low-dimensional structures,^{2,3} which is a consequence of their three-dimensional quantum confinement of carriers and delta-like density of states.

So far, the optical properties of InAs/GaAs QDs have been widely investigated by photoluminescence,⁴ photoluminescence excitation spectroscopy (PLE),⁵ time-resolved photoluminescence,⁶ infrared absorption,⁷ as well as modulation spectroscopy, e.g., contactless electroreflectance⁸ (CER) and photoreflectance (PR).^{9,10} The modulation techniques are known to be nondestructive and very powerful tools for the characterization of semiconductors and their microstructures.^{11,12} The basic idea of the modulation spectroscopy is a very general principle of experimental physics. Instead of measuring the optical reflectance (or transmittance) of the material, the derivative with respect to a modu-

lating parameter is evaluated. It leads to sharp, differential-like features in the spectral region of interband transitions without uninteresting background typically found in common reflectance spectra. In the case of electroreflectance (ER) spectroscopy the electric field in the sample is modulated directly by an ac voltage via electric contacts or without contacts in a capacitorlike system with a semitransparent top electrode. In photoreflectance, instead of directly applying an ac voltage, the sample is perturbed with a chopped laser pump beam. In a first approximation, the modulation mechanism in PR is also considered to be electromodulation (EM), as this is done by most of the authors, and which seems to be indeed justified in many cases. When the laser is on, the photogenerated carriers drift in a built-in electric field and are captured by surface/interface trap states, thus reducing this field. When the laser is off, the trap occupation, and hence the field, is restored. However, PR and ER spectra do not have to be fully equivalent in general, and in certain circumstances photomodulation can lead to another type of modulation related to the existence of extra carriers in the structure. These effects are usually negligible in the case of bulklike or quantum well (QW) structures, because the photogenerated carrier concentration does not change the dielectric properties of the system significantly. However, their influence has been observed previously in a form of the so-called below band gap oscillations, i.e., an interference effect related to the modulation of the refractive index difference between two parts of the structure (e.g., doped substrate and undoped buffer).^{13,14} In the case of a quantum dot structure

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the properties of these nanometer scale objects can be much easier modified due to loading them with charge carriers. In general, the possible modulating factors which may contribute to the QD photoreflectance signal can be divided into two groups: (i) photoinduced electromodulation of the built-in electric fields and hence modulating the QD optical transitions due to quantum confined Stark effect (this factor is assumed to be the dominating one for most of quantum well structures); (ii) modulation of the dot properties due to the appearance of carriers in the QD layer, e.g., self-consistent modification of the confinement potential, many-body effects important at higher carrier concentrations, changes in the dot material refractive index, effect of filling the dot states of a whole inhomogeneous QD ensemble according to the Boltzmann distribution, etc. The second group of effects could be a reason for a modified QD PR signal line shape, making it different than the very well known low field electromodulation functional form.^{11,12}

The aim of the present paper is to study the mechanisms responsible for the PR signal from self-assembled QDs and the modulation origin. We have performed room temperature PR measurements on self-assembled InAs/GaAs QDs designed to have the ground state transition at 1.3 μm suitable for telecommunication applications.^{15,16} Our main experimental approach is to measure photoreflectance excitation (PRE) spectroscopy of QD transitions, i.e., an analogy of photoluminescence excitation. This method has been already successfully applied to quantum well structures or bulklike materials a few times, and it has been proved that it probes the optical transitions in a similar way as PLE and can be used to determine the modulation mechanisms.^{17,18} Here, we demonstrate its applicability for investigation of QD structures as well and we show that the main factor responsible for PR response from self-assembled QDs is the photomodulation of the surface electric field.

II. EXPERIMENTAL DETAILS

The sample investigated here was grown by solid-source molecular beam epitaxy on (001)-oriented GaAs substrates. Two InAs layers with a nominal material amount of about 2.1 monolayers each were deposited on an undoped 0.5 μm thick GaAs buffer to form QDs by self-assembly. The QD layers were separated by 11 nm GaAs spacer (a possible quantum-mechanical coupling between the two layers can be neglected in a first approximation for the aim of this paper). The whole structure was covered with a 100 nm thick GaAs capping layer.

In order to measure PR and CER spectra the so-called bright configuration of PR setup has been used.¹⁹ Light from a tungsten halogen lamp was reflected off the sample and then came through a single-grating 0.55 m focal-length monochromator and was detected by thermoelectrically cooled InGaAs photodiode. The pump beam for PR was provided by the 532 nm line of a frequency doubled Nd:YAG (yttrium aluminum garnet) laser. The laser light was chopped by a mechanical chopper at a frequency of 275 Hz. PRE measurements¹⁷ have been performed using the same setup, in which the detection is set at a constant energy (at QD

FIG. 1. Room temperature PR spectra obtained at the modulation wavelengths of 532 nm (a) and 800 nm (b), and nonresonant high excitation PL spectrum (c).

ground state transition) and the wavelength of the pump beam is changed (light from tunable Ti: sapphire laser) from 800 to 1000 nm. In the case of CER measurements the sample was mounted in a capacitor with the top electrode made of a copper mesh and the bottom electrode made of a copper solid block. The top electrode is semitransparent and was kept at a distance of approximately 0.1 mm from the sample surface while the sample itself was glued to the bottom copper electrode. A maximum peak-to-peak alternating voltage of 1.8 kV was used for the modulation. Phase sensitive detection of PR and CER signals was made using a lock-in amplifier. Similar equipment was also used to measure room temperature photoluminescence (PL) and PLE in standard experimental configurations.

III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show room temperature PR spectra for the InAs/GaAs QD structure measured with pump beam wavelengths of 532 and 800 nm, respectively. The spectra are very similar, i.e., qualitatively independent of the pumping light wavelength (photon energy), for energies higher than main energy gap of the structure. At least three groups of features can be distinguished in these spectra. The most intensive one at the energy of about 1.42 eV is associated with the bulklike band gap transition in GaAs, which is the base material of the whole sample. Just below that, two relatively sharp lines at 1.34 and 1.38 eV are clearly visible. The origin of those can be confirmed when compared to some works reporting on wetting layer (WL) transitions in InAs/GaAs self-assembled QD structures or with the results

FIG. 2. Room temperature PRE and PLE spectra for two different pumping/excitation power densities: (a) 100 mW/cm² and (b) 400 mW/cm² (normalized to the maximum intensity).

of common effective mass envelope function calculations of energy levels in a very narrow and strained InAs/GaAs QW.^{8,9} In this way, the WL1 and WL2 resonances in Fig. 1 can be attributed to the heavy- and light-hole ground state transitions in the WL related QW, respectively. The third group of PR features in the energy range below the WL transitions is associated with optical transitions in InAs/GaAs QDs. The intensity of QD transitions is much weaker than the intensity of WL and GaAs related ones due to low amount of QD material in comparison with InAs wetting layer and GaAs cap/buffer materials (QDs cover only a small fraction of the entire illuminated area of the sample). High excitation photoluminescence spectrum has been measured on the same sample at room temperature [Fig. 1(c)], confirming the origin of the QD-related PR lines.

In order to examine the sources of modulation responsible for PR signal related to an ensemble of QDs, the PRE experiment has been performed, in which the QD ground state PR feature intensity has been analyzed as a function of the energy of the pump beam photons. Figure 2 shows such a dependence versus the tuned wavelength of the pumping laser (tunable Ti:sapphire laser in this case) in the range from energies far above the GaAs band gap down to energies below the WL transitions for two different pump beam power densities. For a comparison, similar dependences for the PL intensity (photoluminescence excitation) have been presented. In general, both should just reflect the absorption spectrum of the sample and should be comparable showing the energies at which an enhanced absorption occurs, but they appeared to be not exactly the same as it is seen in Fig. 2. The PLE spectrum shows indeed when the absorption increases (at which energy or wavelength) what is observed through an increased emission of the QD ground state exci-

ton (if only the photogenerated carriers reach the dot and can contribute to the emission intensity). The PRE spectrum reflects the modulation efficiency, which is also higher when the absorption is stronger, but the photogenerated carriers do not need to reach QDs to contribute to the modulation. It is enough if they contribute to the built-in electric field changes in the sample. And this is the reason for some differences between PLE and PRE spectra. In the former one, it is seen that QD PL intensity decreases rapidly, when only the excitation energy becomes smaller than the GaAs band gap, i.e., the concentration of photogenerated electron-hole pairs drops down drastically. Then, some intensity increase is observed at the energy of the WL ground state transition, i.e., the electrons and holes created in the wetting layer are trapped by the dots and recombine through their ground state (excitonic recombination, in fact). The PLE intensity increase at the WL related part of the spectrum is much weaker than that at the energy above the GaAs band gap, because the absorption in a thin WL layer is much weaker than that in bulklike GaAs layers. In the PRE spectra, however, some QD PR intensity is still detected for pumping energies lower than GaAs band gap, i.e., the PRE spectral edge is shifted to the red in comparison with the PLE one and has a longer low energy tail. It is related to the fact that the illumination of the sample with below band gap photon energy is still causing some internal electric field modulation. In such a case, the carriers originate from defect/dopant states in the gap, i.e., no electron-hole pairs can be created and only one type of additional carriers appears in the structure. Therefore, they can contribute to the PRE signal but they cannot contribute to the exciton emission in the dots. A finite modulation efficiency for below band gap modulation has been also confirmed by the direct observation of the GaAs feature (see Fig. 3) and it was reported previously for various types of structures and semiconductor materials.^{20,21} Additionally, no PRE intensity increase has been observed at WL energy. It shows that the possible modulation is related to the carriers appearing in the dots via the WL states (due to absorption at WL QW heavy-hole ground state transition energy), and probably similarly with those created directly resonantly in the dots is negligible. In general, it is expected that these carriers cannot significantly influence the electric field distribution but they could affect the transition energies or intensities due to any of the charge carrier concentration related effects (also when the carriers are created directly in the dots or via the WL states). It appears to be not the case, because no fingerprints of these effects are seen in our PRE spectra (see Fig. 2). It brings us to the conclusion that the effect responsible for the PR modulation of the signal for an ensemble of dots is dominated by the built-in electric field modulation independently of the pump beam wavelength (energy) thanks to the carriers created in the barrier material (as long as they can be created in the barrier and any signal can be detected). In our case, the dot layers are under influence of the surface electric field (they are placed 100 nm from the surface only). A schematic diagram of the band bending with QD confining potential and its modification due to sample illumination is shown in Fig. 4. Usually, the maximum value of the surface electric field is about several kV/cm, which means that the field in

FIG. 3. Room temperature PR spectra obtained at above and below GaAs band gap modulation.

the dot area is too weak to cause any effects observable directly,²² but its photogenerated changes are strong enough for modulation due to quantum confined Stark effect and are seen in reflectivity coefficient changes.

An additional comment is needed regarding the effect observed in Fig. 2, where some apparent energy shift of the PRE spectrum can be observed when the pumping power is increased, whereas nothing like that has been seen for PLE spectra. The explanation is related to the origin of the efficiency of the modulation for pumping photons with energy below but close to the fundamental GaAs absorption edge. As it has been mentioned above, the below band gap modulation is usually related to the carriers generated from some defect states. But the density of these states can be quite easily saturated at a certain power level and further pump power increase can only cause the above band gap carrier generation. The increase of the PRE signal intensity due to the latter reason will just dominate the entire spectrum for

FIG. 4. Band bending at the surface of InAs/GaAs QD structure. The illuminated situation is also schematically shown.

FIG. 5. Comparison of PRE spectra measured at two different pumping powers presented in the natural (not normalized) scale.

high modulation power densities and the defect related modulation disappears in the low energy tail (see Fig. 5). Therefore, the edge of the PRE spectrum shifts to higher energies with power increase and approaches the edge seen in PLE, because the defect related modulation becomes saturated for high pumping powers.

In order to confirm our PRE results and conclusions based on them, we have performed CER measurements on the same sample. If the modulation mechanism in PR is mainly related to the electromodulation of the surface electric field then the spectra (in the QD part at least) should be similar including also their line shape. Such an agreement has been obtained and demonstrated in Fig. 6. Therefore, it is also an argument for using the well known low-field functional forms^{11,12} to fit the PR data (and extract the QD transition features) obtained for self-assembled dots in structures, where the electromodulation mechanism can be treated as the dominating one.

FIG. 6. Comparison of room temperature PR (a) and CER (b) spectra.

IV. CONCLUSIONS

In conclusion, we have investigated the sources of modulation in photorefectance, which are responsible for the $\Delta R/R$ signal related to an ensemble of self-assembled InAs/GaAs QDs. It has been demonstrated on the basis of a set of photorefectance and photoluminescence experiments, but also PRE, PLE, and CER spectra, that the QD PR signal existence and its intensity can be fully explained assuming that the most effective modulation factor is the photomodulation of GaAs surface field, which via the quantum confined Stark effect causes the modulation of the energy, broadening, and intensity of the QD transitions. Hence, the line shape typical for electromodulated spectra seems to be the most appropriate for fitting the QD features.

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