Defect Creation in InGaAs/GaAs Multiple Quantum Wells – II. Optical Properties

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1. Introduction

High device performance requires optimized growth processes tailored to the underlying epitaxial structures, in an effort to reduce crystalline deterioration as a possible origin for device capability shortcomings [1-3]. In this respect, crystal defects are known to mediate loss mechanisms [4], to be responsible for leakage channels [5], and to be roots of device degradation [6]. In the context of optoelectronic applications, their impact on optical properties has hence been studied extensively for various material systems.

In the InGaAs/GaAs material system, for instance, it was shown that dislocation edge segments particularly enhance non-radiative recombination. The involved carrier states, mediating this loss mechanism were revealed to be associated with dislocation cores, and did not originate from spatially separated defect sites [7].

Our x-ray diffraction investigations [8] previously revealed distinct dependencies of the crystalline properties of InGaAs/GaAs multiple-quantum-well (MQW) structures on molecular beam epitaxy (MBE) growth conditions. In this paper, these findings are related to conclusions drawn from optical studies. In particular, non-radiative recombination is assessed by means of a model developed herein, in order to establish correlations between growth conditions, crystal quality, and optical properties for the particular structures investigated herein.

2. Experimental Procedure

Three sets of $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ MQW samples were grown by MBE on undoped (001)-oriented GaAs wafers. The first set consists of four samples grown at varied deposition temperature 475-520°C, the second set was grown with varied periodicity $\times 5$ and $\times 10$, and finally a third set features a MQW period series of 4+8 nm, 8+8 nm, and 8+16 nm. A full description of the growth conditions and the setup used can be found in the companion paper [8]. In addition to characterization by means of high-resolution x-ray diffraction (HRXRD) described therein, optical properties were examined in various photoluminescence spectroscopy (PL) experiments.

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Information about the electronic states present in the grown structures was extracted from temperature-dependent PL measurements. Placed in a He-flow cryostat between 5 and 300 K, the samples were excited with a 632.8 nm HeNe-laser at 10 mW optical power. The resultant PL transition energies were compared to the calculated confinement states of carriers in these nanostructures obtained using self-consistent Schrödinger Poisson methods [9].

Excitation power-dependent photoluminescence spectroscopy was utilized to characterize the strength of non-radiative recombination in the structures as a result of the defects created during epitaxial growth. The measurements were carried out at 12 K with excitation provided by a 376 nm laser diode and power varied over three orders of magnitude up to 70 mW.

3. Excitation-dependent PL for defect analysis

A model is presented as a means to characterize the strength of Shockley-Read-Hall (SRH)-recombination based on excitation power-dependent PL measurements. Fundamental to this model is a transition of the dominant carrier recombination process from one type to another depending upon the excess carrier density. This manifests as a change in scaling of the light output-excitation power relationship from a square to a linear dependence.

At low temperature and sufficient excitation power, the SRH-recombination rate per unit volume in undoped structures is given as $R_{SRH} = \frac{A}{\tau} \cdot \Delta n$, with the SRH-parameter A and the excess electron density Δn [10]. Optical excitation creates electron-hole-pairs $\Delta n = \Delta p$, (Δp is the excess hole density). In structures without carefully designed optical cavities, spontaneous radiative recombination occurs at a rate $R_{spo} = B \cdot (\Delta n)^2$, with the spontaneous emission coefficient B . A third recombination process, Auger recombination, occurs at the rate $R_{Auger} = C \cdot \Delta n^3$, with the Auger coefficient C ¹. [11]

These recombination mechanisms compete with carrier generation by the excitation source (power P , wavelength λ). The generation rate G per volume V is proportional to the excitation power, $G(\lambda) = \frac{P}{h\nu} \cdot \frac{1 - e^{-\alpha d}}{d}$ where h is Planck's constant, c is the speed of light, α is the absorption coefficient, and d is the length of the absorption region. At steady state, the rate equation for these processes reads $\frac{dn}{dt} = G - R_{SRH} - R_{spo} - R_{Auger} = 0$. Simplifications are possible if Auger recombination is neglected. For that reason a means is introduced to determine from experimental data if this condition is met. The internal quantum efficiency IQE is given by $IQE = \frac{R_{PL}}{R_{PL} + R_{SRH} + R_{spo}}$. Fig. 1 shows an example of the calculated dependence of IQE on carrier density n for selected values A , B , and C . Beyond a plateau within a carrier density range where radiative recombination dominates, a drop in IQE marks the onset of Auger recombination as the dominant mechanism. The external quantum efficiency $EQE \propto IQE$ is experimentally accessible since $EQE \propto \frac{I_{PL}}{P}$, where I_{PL} is the measured PL intensity. Thus a plot of the quantity $\frac{I_{PL}}{P}$ versus the excitation power P allows one to determine the experimentally covered excitation range over which Auger recombination can be neglected. Indirect semiconductors with a low B parameter are not suitable for investigation in this manner.

From the approximated rate equation $0 = G(\lambda) - R_{SRH} - R_{spo}$, in which Auger recombination is neglected, the rate of emitted photons from the optical volume V is derived as

$$I_{PL}(\lambda) = \frac{P}{h\nu} \cdot \frac{1 - e^{-\alpha d}}{d} = \frac{A}{\tau} \cdot \Delta n + B \cdot (\Delta n)^2. \quad (1)$$

¹ In undoped structures held at low temperature the electron density n can be approximated as $n \approx \Delta n$.

Approximation of this equation in low excitation limit ($\eta \ll 1$) and at intermediate level ($\eta \approx 1$), shows square and linear scaling, respectively. A sample plot of Eq. (1) illustrates the two regimes (Fig. 2). At low pumping level, carriers mainly saturate trap states and only a small portion recombine radiatively. With increased pumping levels, non-radiative recombination sites become saturated and radiative recombination of electron-hole-pairs dominates. The transition between the two regimes occurs gradually $\eta PL \propto \eta^2$ with a continuously decreasing scaling parameter $\eta \in 1,2$. A particular value of the power at which the transition occurs can be defined as the excitation level G_T at which the curves, approximated in the individual limits, intersect. Since the generation rate at this point is given by $\eta T = \eta^2 G \propto \eta^2$, the excitation power at which this transition occurs can be used to measure the magnitude of the A parameter.

To summarize, excitation power-dependent PL measurements can be used to detect a transition from square to linear scaling in the relation between the intensity of the luminescence emitted from the sample and the excitation power. The higher the excitation power at which this transition occurs, the stronger are the non-radiative processes. Applying this ABG-model allows one to draw conclusions about how defects created in epitaxial growth act as non-radiative recombination sites.

4. Results

The quantum confinement states of carriers in MQW nanostructures are studied using PL over a range of temperatures and excitation powers. The temperature evolution of the PL transition lines reveals the nature and origin of these states. Ideal behavior would reproduce the temperature-dependent transition energy behavior between electron and hole confinement states.

4.1 Effect of different deposition temperature

As described in the companion paper [8], one sample set consists of $\times 5$ MQW structures among which the deposition temperature was varied. Temperature-dependent PL revealed Varshni-like temperature dependence of the main PL transition line for all samples grown at $T_{\text{dep}} \leq 505^\circ\text{C}$ [14]. An example of the PL peak dependence on ambient temperature for the structure with $T_{\text{dep}} = 505^\circ\text{C}$ is shown in Fig. 3a. Good agreement with the calculated transition energy between heavy hole and electron MQW states (black, dashed) shows ideal carrier confinement in the nanostructures. The low temperature (12 K) linewidth of the PL peaks were as low as 8 meV, indicating a low density of defect centers at which carriers are scattered, which would otherwise lead to PL line broadening. This range of deposition temperatures ($\leq 505^\circ\text{C}$) resulted in high quality $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ MQW structures with ideal carrier confinement.

The method described in Sec. 3 is applied to three samples with $T_{\text{dep}} \leq 505^\circ\text{C}$. The ratio $\eta PL \propto \text{EQE}$ vs. P is plotted in Fig. 4 to validate the assumption of negligible Auger recombination. It is apparent that the regime in which Auger recombination becomes significant is not entered, i.e. the EQE does not drop beyond the plateau as it would at higher excitation as shown in Fig 1. Hence the simplified equations of the ABG-model (Eq. 1) can be used for further analysis.

The change in the scaling parameter k of an I_{PL} vs. P curve is a measure of the SRH-recombination strength. A suitable way to present this transition is shown in Fig. 5, in which the scaling exponent $\eta = d \log(\eta PL) / d \log(\eta)$ is plotted against the excitation power P . While square scaling corresponds to $k=2$, linear scaling is indicated by $k=1$. The plot of the scaling exponent shows different onsets of linear scaling over the covered excitation range. The structure $T_{\text{dep}} = 505^\circ\text{C}$ exhibits linear scaling over the entire excitation range as the scaling exponent scatters around 1. For the structure grown at 15°C lower temperature the scaling exponent started off around 1.2 at the lowest pumping level, decreasing continuously with excitation power and finally reaching unity. In comparison to the structure grown at

higher temperature, this sample required a higher excitation power to reach the linear regime. The lowest deposition temperature sample $T_{\text{dep}}=475^{\circ}\text{C}$ manifested a high scaling exponent ~ 1.4 at lowest excitation. Although k decreased continuously with higher excitation, linear scaling was not observed in the covered range. The value $k=1$ is hence likely to occur at even higher pumping. These measurements showed a gradual increase of the pumping level at which the linear PL regime is entered as deposition temperature is decreased, leading to the conclusion that the strength of non-radiative SRH-recombination increases likewise.

Whereas the structures with $T_{\text{dep}} \leq 505^{\circ}\text{C}$ showed Varshni-like behavior in the temperature-dependent PL experiments as a result of ideal carrier confinement, the sample with $T_{\text{dep}}=520^{\circ}\text{C}$ deviates in this respect (Fig. 3a). In contrast to the simulated transition energy dependence (black, dashed), this sample's PL response (grey) started off with a photon energy ~ 50 meV below the calculated value at low temperature and revealed a non-monotonic temperature dependence. This particular S-shape behavior and the non-monotonic variation in peak asymmetry (compare peak and central wavelength) hint at the localization of carriers in local energy minima. Redistribution of carriers among local minima is a temperature activated process and leads to the S-shape dependence observed here. Along with the non-monotonic linewidth behavior and a good fit of the temperature-dependent PL intensity data to a model proposed by [15, 16] (both Fig. 3b) the results suggest the occurrence of carrier localization. This mechanism has been described previously in various other structures [15-17]. The identified presence of random band edge fluctuations is attributed to compound composition variations as a result of InGaAs decomposition at an elevated deposition temperature $T_{\text{dep}}=520^{\circ}\text{C}$.

4.2 Effect of MQW periodicity

This set of samples probes the influence of MQW periodicity on the optical properties of epitaxial structures. The PL response showed good carrier confinement in these epitaxially grown nanostructures with agreement between measured photon wavelength and calculated confinement states. To compare the influence of defect related recombination processes, excitation-dependent PL was carried out according to the ABG-model used above. As was shown above for the temperature series (Fig. 4), the behavior of the quantity η_{PL} was examined with regard to a possible EQE drop at highest pumping level, justifying neglecting Auger recombination in the ABG-model. The scaling exponent dependence on the pumping level is plotted in Fig. 6 for the samples with different MQW periodicities. It can be seen that the low-periodicity structure revealed a scaling exponent ~ 1.2 at the lowest excitation and rapidly transitioned into the linear region. In contrast, the high-periodicity sample started off with a higher scaling exponent ~ 1.9 and reached linear scaling at a comparatively higher pumping level. Based on the ABG-model it can be inferred that trap state mediated non-radiative recombination is stronger in the $\times 10$ structure. An increase in the periodicity of epitaxial MQW structures led to a reduced carrier lifetime as a result of more defect sites involved in SRH-recombination.

4.3 Effect of MQW period

The decrease of the MQW period from $8+16$ nm to $8+8$ nm of epitaxial structures is supposed to result in different densities of defects due to the different amount of total elastic strain.

As before, the strength of SRH-recombination was investigated via excitation power-dependent PL. The evolution of the ratio η_{PL} confirmed that Auger recombination is negligible over the excitation power range of interest. A plot of the scaling exponent versus the pumping level is shown in Fig. 7. It is apparent that the sample with highest MQW period $8+16$ nm stays in a linear regime over the entire

excitation range. The saturation of recombination sites in the epitaxial volume is expected to occur at even lower excitation power. Reducing the MQW period to 8+8 nm reveals the higher pumping level necessary to enter the linear scaling regime.

Hence, the reduction of unstrained barrier layer thickness from 16 to 8 nm increased the strength of SRH-recombination considerably. This result, however, has to be put into perspective, as the limited absorption depth (~ 200 nm [18]) leads to partial excitation and hence examination of only part of the MQW volume of the structure with MQW period 8+16 nm. The substrate/epi-structure interface, which due to relaxation of the initial elastic stress [8] is a probable location of non-radiative recombination sites, most likely contributes only slightly to the emitted light and is thus of minor influence in this ABG-experiment because of the sample's thickness ($\times 10$, 8+16 nm). If the scaling exponent curves of this structure and the $\times 5$, 8+8 nm one (the latter revealing comparatively reduced edge segment density and no relaxation) are compared, it becomes apparent that non-radiative recombination is underestimated in the thick structure 8+16 nm.

5. Correlation with crystalline properties

The optical properties of various InGaAs/GaAs MQW structures studied in this work were correlated with the particular growth conditions and crystalline properties from prior work as explained in detail in [8].

In the set of samples grown at varied deposition temperatures, HRXRD investigations found an optimal growth temperature $T_{\text{dep}}=505^\circ\text{C}$ for $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$, characterized by smooth MQW interfaces, minimal dislocation loop (DLs) densities and the least amount of diffuse scattering in symmetric (004) reciprocal space maps. At this value and below ($T_{\text{dep}} \leq 505^\circ\text{C}$) the main type of crystalline defects are 60° DLs with increasing density as T_{dep} is decreased. From the optical data, the primary and secondary DLs at the measured densities ($3.0\text{-}5.5 \times 10^5 \text{ cm}^{-2}$ and $1.0\text{-}2.7 \times 10^7 \text{ cm}^{-2}$) still allow proper confinement of charge carriers in the nanostructures as concluded from temperature-dependent PL experiments. The strength of non-radiative SRH-recombination increases along with the density of DLs for lowered T_{dep} and the optimal T_{dep} found in HRXRD experiments coincides with the one inferred from optical investigations.

Exceeding the optimal $T_{\text{dep}}=505^\circ\text{C}$ revealed a material instability manifested in InGaAs decomposition and a structural instability visible as lateral layer thickness undulations in transmission electron micrographs. Both deteriorations cause local effective band gap fluctuations as both the compound composition and confining layer thicknesses determine carrier state energies. This correlates with random band edge fluctuations found in the optical investigations (Fig. 3) that led to carrier localization in local energy minima.

Varied MQW periodicity from $\times 5$ to $\times 10$ at the same T_{dep} and MQW period revealed reduced vertical and lateral coherence, an increased density of secondary 60° DLs, and visible relaxation of initial elastic stress up to $\sim 15\%$. As the densities of edge segments on the substrate/epi-structure interface and Lomer dislocations in the volume increase non-radiative SRH-recombination becomes prominently stronger. This finding was explained as a cumulative deterioration effect. Denser creation of edge segments close to substrate/epi-structure interface (tertiary DLs) giving rise to stress relaxation [19-21] accounts for the denser distribution of defect states involved in SRH-recombination.

Lastly, a set with different MQW periods, grown at the same T_{dep} with common MQW periodicity, showed in its optical properties the effect of thicker unstrained barriers as defect “healing” layers while the period was increased from 8+8 nm (well + barrier width) to 8+16 nm, in agreement with the conclusions drawn from HRXRD investigations.

6. Discussion and Conclusions

In this work, a method capable of evaluating the strength of defect state mediated non-radiative recombination was introduced. This ABG-model is based on excitation power-dependent PL experiments. Its applicability was shown for several InGaAs/GaAs MQW structures. Limitations of this model were observed for structures with weak carrier confinement, such as shallow wells (e.g. GaAs/GaAs_{0.92}Sb_{0.08}) or comparatively thick single layers (e.g. 100-500 nm), based on data not presented herein. However, the proposed ABG-model can be applied to nanostructures with proper carrier confinement, as typical for, e.g. light emitting diodes and MQW solar cell structures.

Further, correlations between the crystalline and optical properties versus epitaxial growth conditions were presented. Investigation of three individual sample sets with variations in deposition temperature, total MQW thickness, and MQW period, allowed the creation of extended and optically active defects to be studied systematically.

The correlation between physical properties, such as the optical response and crystal perfection, as dependent on growth conditions are of special importance in the development of growth processes for device applications. In this context, optimization of the growth procedure and prevention of material quality deterioration improve device performance.

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All figures single column fitting images.

Fig. 1: Calculated internal quantum efficiency (IQE) based on a conservative low temperature life time estimate and appropriate B and C parameters [12, 13]. Until the onset of the IQE drop at elevated carrier density n , Auger recombination can be neglected.

Fig. 2: Sample plot of the emitted PL intensity upon increase of the excitation power. Two distinct regimes, characterized by square and linear scaling, respectively are identified. The transition occurs at a higher pumping level the stronger SRH-recombination.

Fig. 3: a) Temperature-dependent PL signal for two MQW structures with the same periodicity and period. Whereas the structure grown at $T_{\text{dep}}=505^{\circ}\text{C}$ reveals a behavior that follows closely the calculated trend (black, dashed), sample $T_{\text{dep}}=520^{\circ}\text{C}$ is described by an S-shape curve. b) Characteristic non-monotonic linewidth temperature dependence and good fit of integrated PL intensity versus ambient temperature curve to $\Phi\text{PL}=\Phi_0(1+\Phi\cdot\exp(\Phi\Phi_0))$ suggest the occurrence of carrier localization.

Fig. 4: The shape of the external quantum efficiency (EQE) curve as dependent on excitation power indicates that Auger recombination is negligible over the covered pumping range, if compared to Fig. 1.

Fig. 5: Linear scaling occurs at a higher excitation power the lower the deposition temperature T_{dep} . High scattering of data points is to be expected as the plotted quantity (scaling exponent) is based on a differentiating method.

Fig. 6: Direct comparison of optical behavior at varied excitation power unveils the stronger SRH-recombination in the higher periodicity MQW.

Fig. 7: Increasing the unstrained barrier width reduces defect state mediated non-radiative recombination as apparent from particular scaling exponent behavior shown.

Figure 1