In-depth and in-plane profiling of light emission properties from semiconductor-based heterostructures

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Cathodoluminescence (CL) technique is applied for evaluation of in-depth and in-plane variations of light emission from semiconductor heterostructures, including laser diode structures. Light emission properties of heteroepitaxial and homoepitaxial structures are studied. We demonstrate possibility of in-depth profiling of complicated multi quantum well structures, which allows us to evaluate light emission characteristics from different regions of, e.g., laser structures. Due to this property of the CL, we can evaluate interconnections between structural quality of the samples and light emission characteristics. Stimulated emission under electron beam pumping is achieved in a conventional CL set up for selected heterostructures. Threshold currents for stimulated emission are evaluated from the CL investigations. We demonstrate that potential fluctuations are not fully screened in the active regions of laser structures, even at large excitation densities.

Keywords: semiconductors, heterostructures, cathodoluminescence, depth-profiling, defect distribution, laser emission.

1. Introduction

A wide spread use of semiconductor materials in electronic and opto-electronic devices promotes detail studies of their properties. Most of available experimental techniques averages material properties, making difficult to conclude on in-plane and in-depth distribution of defects and impurities. In this work, we demonstrate that such information can be obtained from scanning, spot-mode and in depth-profiling cathodoluminescence (CL) investigations. We show that in-plane and in-depth properties of semiconductor quantum well (QW) heterostructures can be studied with atomic-like resolution, which enables direct observation of light emission from active regions of semiconductor-based devices, as demonstrated for GaN-based laser diode (LD) structure.

2. Experimental

The CL spectra were taken in a JEOL35C scanning electron microscope with a MonoCL2 CL system by Oxford Instruments, using a 1200 lines/mm grating blazed at 500 nm and detected using a Hamamatsu R943-02 Peltier cooled photomultiplier. Spectra were not corrected for the system’s response. The electron beam current \( I_b \) was measured using a Faraday cup. All spectra discussed here were taken at room temperature. Charging effects were carefully minimized.

3. Basics of CL depth-profiling experiments

Resolution of CL investigations is not that of focused beam of primary electrons, but depends on a radius of a scattered cloud of primary and secondary electrons and on a diffusion length of carriers or excitons [1–3]. Information on in-plane emission properties, obtained by scanning electron beam through a given area, is thus limited to at least 50 to 100 nm [3]. In-depth resolution is available through CL investigations taken with varying beam energy (accelerating voltage) [1,2,4–7]. Interpretation of the depth profiling CL data is based on a theory of CL depth-profiling experiments shortly described below. Empirical function describing

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electron generation rate \( G(z) \), with \( z \) normal to the surface of the sample, is given by \([2, 5, 8]\)

\[
G(z) = \frac{1}{R} 0.6 + 6.21 \left( \frac{z}{R} \right) - 12.4 \left( \frac{z}{R} \right)^2 + 5.69 \left( \frac{z}{R} \right)^3, \quad (1')
\]

for \( 0 \leq z \leq 1.1R \), and

\[
G(z) = 0 \quad (1'')
\]

for \( z > 1.1R \), where \( R \) (primary electron range) is given by the empirical function

\[
R = (0.052/\rho)E_{\text{beam}}^{1.75}, \quad (2)
\]

with \( R \) in \( \mu m \), \( \rho \) (material density) in \( g/cm^3 \) and \( E_{\text{beam}} \) (electron beam energy) in keV [9]. More advanced analysis requires Monte-Carlo simulations to describe accurately electron trajectories in CL experiments [10].

4. Depth-profiling investigations of CdTe-based QW structures

Using Eqs. (1) and (2) we can estimate the penetration range of primary electrons [Fig. 1(a)] and profiles of electron generation in CdTe. The latter are shown in Fig. 1(b) normalized to an unity. Both curves were calculated for beam energies used in the present study. We assumed here that the penetration ranges are not significantly affected by the presence of two different materials (QW and barrier) of slightly different density. We also assumed that the presence of interface is not important. Below we show that these assumptions result in some overestimation of penetration lengths of primary electrons.

Depth-profiling investigations were performed in two options – at constant excitation power or at constant excitation current. Both gave similar in-depth resolution. Thus, most of the data shown below are those taken at constant current conditions. The experiments were performed for two CdTe-based multiple QW structures grown by molecular beam epitaxy (MBE). The first sample consisted of five CdTe QWs [4, 8, 12, 26, and 32 monolayer (ML) wide] separated by 125 ML wide CdMnMgTe barriers with 15% of Mn and 10% of Mg. The second consisted of four CdTe QWs (2, 4, 6, and 10 ML wide) separated by 50 nm wide CdMnTe barriers with 68% of Mn and with 100 nm wide CdMnTe cap layer.

In Fig. 2, we show room temperature (RT) CL spectrum of the first QW structure. CL emission from all five QWs is...
well resolved even at RT, with relative intensity of CL from different QWs depending on excitation conditions, as is shown in Fig. 3, showing depth-profiling CL data taken between 5 and 12 kV accelerating voltages (between 5 and 12 keV energies of primary electrons). Figure 4 summarizes the results of the depth-profiling CL investigations, proving possibility of optimization of excitation conditions for a given QW, and indicating that in-depth resolution of experiment can be achieved.

Even better in-depth resolution was achieved for the second sample, showing much stronger disorder in photoluminescence investigations [11,12]. For this sample, the strongest emission from the upper most QW (2 nm wide) is achieved at 7 kV accelerating voltage, at 8 kV for the 4 nm wide QW, at 10 kV for the 6 nm wide QW, and finally at about 13 kV for the deepest, 10 nm wide CdTe QW. Comparison of these accelerating voltages with those predicted from Eqs. (1) and (2) indicates that we slightly overestimate electron penetration ranges in the data shown in Figs. 1(a) and 1(b). This partly is due to enhanced scattering at interfaces and also due to the fact that QW and barrier regions differ in a material density.

Important consequence of the depth-profiling CL study is the possibility of optimization of excitation conditions to get dominating emission from a given region of a QW structure. In the consequence, we can study in-plane properties of emission from a given QW and compare them to similar data taken from the same region of the sample at conditions optimized for excitation of other QWs. Examples of such investigations are shown in two images in Fig. 6. These two images show in-plane changes of QW emission for the 2-nm and 4-nm wide QWs for the CdTe/CdMnTe sample. The scanning CL spectra were taken at 7 kV accelerating voltage and at 5000 magnification, with detection set at given QW emission while scanning exciting e-beam through a selected region on a sample. For both QWs the CL emissions vary significantly, depending on excitation spot. The observed white and black patterns often correlate, suggesting the same origin of the observed CL variations.

To support this observation, we performed spot-mode CL investigations. In this study we measured the CL spectrum containing emission from all four QWs, selecting three different white and black spots from the area shown...
in Fig. 6. The data shown in Fig. 7 were normalized to a constant intensity of the CL emission from the 4-nm wide QW. The so-normalized CL emissions vary in an identical way for three wider, and deeper from the surface, QWs, but not for the 2-nm QW, i.e., the one closest to the surface. This CL emission shows much larger intensity changes, suggesting that the dominant factor affecting the observed emission changes are surface related properties. Dislocations present there, irregularities in dopant/contaminant concentrations, height differences, etc... all contribute to the observed in-plane CL intensity changes.

5. Depth-profiling investigations of doped GaN epilayers

GaN is the most promising material for short wavelength opto-electronic applications [13,14]. Very bright green, blue, violet, and ultraviolet light emitting diodes were constructed based on InGaN/GaN/AlGaN heterostructures. Surprisingly, our knowledge of light emission properties of these structures is still inadequate [14,15]. Several studies indicate that we must understand micro-structure properties of these heterostructures to explain mechanism of radiative recombination [14,15]. Scanning CL investigations indicated crucial role of localization processes, related either to micro-structure of the films [3], or indium fraction fluctuations in InGaN QWs [16].

Our present depth-profiling CL investigations indicate that a characteristic property of doped GaN layers are also large in-depth variations of impurity density. Figures 8(a) and 8(b) show in-depth variations of intensity of band edge emission (a) and yellow CL emission (b). Both these emissions are defect-related. Band-edge emission is dominated by bound excitonic transitions, whereas yellow emission is of donor-acceptor pair (DAP) transitions origin [14]. Figure 8(a) shows that “edge” emission comes from upper most layer of the film and gradually decreases in intensity towards an interface to sapphire (GaN sample studied was grown by metalorganic vapour phase epitaxy (MOVPE) on sapphire substrate). A different in-depth property is observed for the yellow emission. This emission is often enhanced at interface region, which apparently is impurity decorated. The latter observation is supported by the data shown in Fig. 9 for the p-type MOVPE-grown GaN sample showing another DAP-related emission at about 3.2 eV [14]. This DAP emission is slightly enhanced at surface...
close region of the sample, becomes weaker in depth of the sample, but then, at increased accelerating voltages, rises in intensity and is the strongest at the interface region.

6. Stimulated emission of GaN/InGaN heterostructures under e-beam pumping

Potential fluctuations, resulting in a strong localization effects, are responsible for the efficient light emission from GaN-based light emitting opto-electronic structures [13–15]. These potential fluctuations are assumed to be screened in laser diodes, explaining difficulties in achieving efficient laser emission from InGaN/GaN heterostructures with large dislocation densities [3,13]. The latter assumption can be tested by studying light emission properties of GaN epilayers and structures under electron beam pumping [17].

Basov and coworkers demonstrated (for liquid xenon) laser radiation under an electron beam (e-beam) pumping [18]. The method was then applied to ZnCdSe/ZnSe heterostructures to achieve a blue-colour stimulated emission [19,20], which allowed to avoid limitations due to p-type doping of ZnSe [19]. Doping of the active layer of laser, which reduces the threshold current, is essential only in the case of short nonradiative lifetimes in the device [20]. Otherwise, undoped structures can be used in e-beam pumped structures, as we also done for InGaN QW structures [17].

In the first step we optimized excitation conditions by performing depth-profiling investigations. This allowed us to select the best conditions to excite the CL emission from the active region of homoepitaxial QW structures and to study in-plane properties of a stimulated emission. Excitation density dependence was then measured. Figure 10 shows such in-plane emission properties for the LD structure with an active region consisting of 20 InGaN QWs embedded between GaN barriers. Indium fraction in QWs was about 2%. Width of QWs and of barrier regions was 2.8 nm (QW)/5.3 nm (barrier). Structure was grown on GaN bulk substrate covered with 0.55-µm thick GaN buffer layer.

Strong variations of the CL intensity are observed in scanning CL study, even for excitation densities above the threshold for the stimulated emission. The observed variations of the CL emission directly reflect growth details. Atomic size growth steps are observed in the scanning CL study. The observed fluctuations reflect microstructure of the samples and not indium fraction fluctuations, which typically are of a much smaller scale [16]. In additions to the growth steps we also observed dark spots in regions of dislocations. Importantly, the presence of dislocations is not “killing” a stimulated emission in a macro-scale, and only results in a micro-scale dark spots in the scanning CL spectra.

![Fig. 10. In-plane intensity changes of a stimulated emission for InGaN (QW)/GaN (barrier) homoepitaxial structure measured at 10 kV and at 10000× magnification.](image)

![Fig. 11. E-beam current dependence of InGaN QW emission in homoepitaxial heterostructure measured at room temperature and at accelerating voltage optimized for QW excitation.](image)

![Fig. 12. In-plane intensity changes of a stimulated emission measured for MOVPE-grown InGaN/GaN LD heterostructure at 6000× magnification, 6.7 nA excitation density and 30 kV accelerating voltage.](image)
Further studied were performed for a homoepitaxial LD structure with a cleaved LD cavity of $L = 300 \mu m$ lengths. This LD structure was used previously for optical pumping experiments, discussed in the reference [21], showing record low threshold for the laser emission. Once optimal conditions (accelerating voltage) for the excitation were optimized, threshold current density to excite a stimulated emission could be determined. A clear threshold dependence of the emission is observed. First, the QW emission increases nonlinearly, but then this increase is extremely rapid and is described by a power low dependence, with two characteristic slopes, suggesting change of an emission mechanism for the excitation current larger than 10 nA.

Scanning CL spectrum (Fig. 12), measured with the detection set at stimulated emission, once more shows growth steps, together with relatively few dark spots, which are most likely dislocation-related. Spot-mode CL experiments (Figs. 13 and 14) shows that CL is quenched in the regions of these dark spots (Fig. 13). In addition to the intensity variations we observed relatively large spectral shifts of the stimulated emission, by about 20 meV. These shifts reflect different localization conditions in a QW planes, indium fraction variations, different strain conditions and likely different local electric field values. All these effects are apparently still affecting emission, even at large excitation densities required to excite a stimulated emission.

7. Conclusions

We demonstrate that depth-profiling CL investigations allow for evaluation of different in-depth properties of the samples studied. Influence of growth details, impurity/defect distribution or emission properties from active regions of the devices can be studied, as we demonstrate for two types of heterostructures – CdTe-based and GaN-based. In the latter case stimulated emission under e-beam pumping could be achieved, allowing us to evaluate the role of localization effects and dislocations in the CL emission at relatively high excitation densities.

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References


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