Systematic photoluminescence and electroluminescence study of high-efficiency surface-textured thin-film light-emitting structures.

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ABSTRACT

Very high efficiency GaAs light-emitting diodes are based on surface-textured thin film structures. The technique relies on surface texturing by “natural lithography”, where a monolayer of randomly positioned polystyrene spheres acts as a mask for etching a random diffraction grating. We present results of a systematic experimental study on the influence of the surface-texturing parameters on the efficiency of these LEDs. The study was performed on GaAs/AlGaAs structures optimized for photoluminescence and electroluminescence, respectively. It shows that the maximum enhancement of the light output occurs for spheres of 200 nm to 300 nm diameter, which must cover more than 50% of the surface. The optimum etching depth is approximately 160 nm. Using these conditions, an external quantum efficiency (corrected for the respective emission wavelength) for MBE-grown GaAs light emitting diodes of 10% was achieved for a device of only $50 \times 75 \mu\text{m}^2$ in size.

Keywords: surface texturing, non-resonant cavity light emitting diode, high efficiency light-emitting diode, epitaxial lift-off

1. INTRODUCTION

High efficiency surface-emitting devices are key elements of many optoelectronic systems, e.g. optical interconnects and displays. Light-emitting diodes (LEDs) have the advantage of large production tolerances compared to vertical-cavity surface-emitting lasers and micro-cavity LEDs. However, the efficiency of ordinary LEDs is limited by the small escape cone that the light has when transiting from a higher to lower refractive index interface as in the case of the semiconductor-air interface. This problem can be overcome by surface texturing and the application of a back mirror¹. These techniques result in an enhancement of the light extraction, which can be explained by angular randomization of the photon trajectories caused by surface scattering.

The surface of these LEDs is textured by dry etching with a mask of a randomly packed monolayer of polystyrene spheres (“beads”), a process often referred to as “natural lithography”². In order to maximize the efficiency of surface-textured LEDs, we performed a systematic study on the influence of bead-size, etch depth and the fraction of the LED surface covered by beads on the enhancement of the light output of the semiconductor structure.

This study was first performed on structures optimized for photoluminescence (PL). An increase in the PL signal can be caused by the increased absorption of the pump beam or by the improved extraction of the luminescence or a combination of the two. In order to separate these two contributions, the influence of

the surface texturing on the absorption was investigated on detector structures which were identical to the PL samples except for the doping profile. The results of this study are described in the first part of this paper.

The most promising conditions were then applied to the LED structures. An increase in the efficiency by a factor of six or more could be obtained for various texturing conditions and for various backside mirrors. These results are discussed in the second part.

2. THE PHOTOLUMINESCENCE STUDY

Sample preparation

All the samples were grown by molecular beam epitaxy (MBE). An initial 50 nm layer of AlAs followed the buffer on all the samples, and was specifically grown for the epitaxial lift-off³. The PL structure consists of an n-doped 200 nm thick GaAs layer embedded in between two AlGaAs cladding layers. The total layer thickness excluding the buffer is 2 μm . The processing of the samples began with the “natural lithography” which was performed using polystyrene spheres with diameters between 50 nm and 1.5 μm . Following this, the samples were etched to depths between 100 nm and 600 nm using chemically assisted ion beam etching. At these etching depths, only the top cladding of the PL samples was etched, leaving the active GaAs layer untouched. Fig. 1 shows a typical cross-section of a surface textured by this method.

Fig. 1: Typical cross-section of a GaAs surface textured by chemically assisted ion beam etching with a mask created by natural lithography. During etching, cylindrical pillars are formed. Their height and their diameter correspond to the etching depth and the sphere diameter, respectively.

After removal of the beads, epitaxial lift-off was performed on small pieces of the sample: The textured surface was protected by bees wax, while the sacrificial AlAs layer was etched laterally by HF. After epitaxial lift-off, the wax side of the sample was glued onto a holder using standard photoresist, and loaded into an evaporation chamber. A mirror of 200 nm SiO_2 , 20 nm Ni and 200 nm Au was deposited by e-beam evaporation. After unloading the sample, the bottom side of the mirror was glued on a glass plate using optical glue, and the resist and wax were removed by acetone and trichloroethane, respectively.

Epitaxial lift-off was also performed on a non-textured reference sample from the same growth, which was glued on a glass plate with optical glue without application of a back mirror.

The evaporation of a mirror is an alternate technique to Van-der-Waals⁴ bonding of the epitaxial layer onto a mirror. The 20 nm nickel layer was introduced in order to improve the adhesion of the gold onto the SiO_2 , but led to the reduction of the mirror reflectivity by approximately 25%.

Experimental results

The photoluminescence in the GaAs layer was pumped by a 785 nm laser diode, chosen such that there was no absorption in the AlGaAs cladding layers. The integrated PL signal of the textured samples with an applied back mirror was normalized to the reference sample without a back mirror. The reference sample had its substrate removed in order to avoid the influence of substrate PL on the signal. The results of these measurements are shown in figures 2, 3 and 4.

Fig. 2: Enhancement of the PL signal by texturing the surface with natural lithography using various bead sizes. The etching depth was identically $160 \text{ nm} \pm 15 \text{ nm}$ for all samples. The signal is normalized to a reference sample with a non-textured surface without a backside mirror.

Fig. 3: Enhancement of the PL signal versus etching depth for three bead sizes. The same normalization was applied as in Fig. 2.

Fig. 4: Enhancement of the PL signal versus the fraction of the sample surface covered with beads. The samples were etched together in the same run. As the etching depth is influenced by the coverage factor, it is not precisely identical for all samples. The error bars in the etching depth arise out of a variation of the depth with the position on the sample, probably correlated to the statistical non-uniformity of the coverage.

The results clearly indicate a maximum in the enhancement of the PL signal for a bead size around 200 to 300 nm. The enhancement increases with increasing etch depth, and saturates at depths above 300 nm. There appears to be a tendency for the bead size to become less relevant for larger etching depths.

As the number of scattering centers reduces, a reduction in the scattering effect is expected. As shown in Fig. 4, this behavior was also observed. The systematic variation in the etch depth due to the variation in the coverage factor does not appear to influence this observation: The etch depth of the samples with lower coverage was slightly larger, thus leading to an increase of the signal (see Fig. 3). Since the signal decreases, we can safely conclude that the influence of the etching depth is over-compensated by the smaller number of scatterers.

The data shown in Fig. 4 is representative of packing densities with the largest density being 60%. In principle, the coverage of a monolayer of spheres can reach 90%. However, we could only achieve a maximum density of 70% in the case of bead sizes other than those shown in Fig. 4. For the 70% density, we did not observe a further enhancement of the PL signal. We thus conclude that to obtain maximum efficiencies the optimum coverage factor is between 55% and 70%. Coverages within this range are easy to obtain by the process of natural lithography, which implies that the packing density is not a critical parameter. A coverage of around 60% was applied in all the other experiments presented in this paper.

The reason for the error bars on the PL signals in figures 2, 3 and 4 is the statistical deviation of the signal at various positions of the sample. This deviation in the data is either due to a variation in the layer quality or in the total layer thickness as a function of the position on the sample. Although the surface of the samples was textured, the presence of Fabry-Perot oscillations in the data could still be observed, the implications and source of which is discussed in more details later in the text (see Fig. 8). However, due to the width of the PL spectrum (FWHM ~ 30 nm), this should not have an effect on the luminescence efficiency, as the Fabry-Perot oscillations are averaged out by the linewidth of the spectrum. Since the linewidth of the pump beam was much narrower, the total absorption of the pump-beam is influenced by the thickness of the structure thus influencing the luminescence efficiency.

Interpretation of the PL results

The PL signal is influenced by both the absorption of the pump-beam and the extraction efficiency of the PL, assuming constant internal PL efficiency. These two parameters have been measured simultaneously in the PL experiments. In order to distinguish their influence, a similar study has been performed on surface-roughened detectors^{5,6}.

The detector and PL structures are identical in all respects, the only difference being the doping distribution, i.e. for the detector structure the GaAs layer is the undoped layer in a p-i-n structure. For this type of structure, it can be expected that the photocurrent is a direct measure of the absorption in the GaAs layer.

Following the definition of the detector devices, an identical procedure as that of the PL samples was applied - surface texturing, epitaxial lift-off, evaporation of a mirror and gluing to a glass plate. The photocurrent of these devices was measured as a function of wavelength and compared with a non-textured reference sample without a back mirror, i.e. still attached to its substrate.

Fig. 5: Enhancement of the photocurrent by surface texturing and application of a back mirror compared to a non-textured reference device without back mirror for a wavelength of 785 nm. The etching depth for all samples is 160 nm.

In order to calculate the impact of the enhanced absorption on the enhancement of the PL signal, the photocurrent measurements were evaluated at the same pump wavelength as used in the PL experiment, i.e. 785 nm. In this case, the photocurrent was averaged over a period of the above mentioned Fabry-Perot oscillations in order to eliminate their influence on the data, and compared with the corresponding data of the reference sample. The result is shown in Fig. 5 which qualitatively exhibits the same behavior as the PL measurements. However, the maximum enhancement of the absorption (which is dependent on the absorption coefficient⁵) by a factor of 1.65 in this wavelength range is much smaller than the enhancement observed for the PL samples. Thus, it is clear that most of the enhancement of the PL signal can be attributed to the increase in the extraction efficiency of the luminescence from the GaAs, which is essential for the production of high efficiency LEDs.

In Fig. 6 the extraction enhancement calculated from the data of figures 2 and 5 is shown. It exhibits a broad maximum of the extraction enhancement for bead sizes between 200 nm and 400 nm. The error bars are omitted for simplicity, though an additional small error might be involved due to tolerances in the MBE growth of the two samples which are compared.

Fig. 6: Enhancement of the light extraction due to surface texturing and application of a back mirror. The data have been obtained by correcting the PL enhancement (Fig. 2) for the absorption enhancement of the pump-beam (Fig. 5).

The influence of the etching depth on the photocurrent was also investigated as represented in Fig. 7 indicating a similar behavior as found for the PL (Fig. 4): The absorption increases strongly with the height of the pillars formed by surface texturing. Again, for reasons of simplicity, the error bars on the etching depth are not shown and an error of 20% could be expected for each measured sample. A comparison of the enhancement of both the photocurrent and PL data as a function of etch depth indicates that up to an etching depth of 160 nm the extraction enhancement increases up to a factor of 4. For increasing depths only a slight increase is obtained with a maximum extraction enhancement of 4.5 which was calculated for a 350 nm etching depth.

Fig. 7: Enhancement of the absorption in dependence on the etching depth. The results for roughened samples with back mirror were compared to the non-roughened reference sample. An etching depth of 0 means no roughening.

Obviously, the influence of bead size and etching depth, which define the pillar dimensions on the textured surface, is qualitatively identical for both, absorption and extraction of light. This is expected, as both phenomena depend on the degree of randomization, which occurs when the light is reflected at the surface. The higher the scattering at the surface, the higher the degree of angular randomization of light inserted into the semiconductor⁵, which enhances the absorption. On the other hand, a higher scattering efficiency results in a smaller number of passes that the light has to undergo in the given material, before it can escape; thus reducing the parasitic absorption and enhancing the light output.

In conclusion, an optimum angular randomization is obtained for pillar diameters which are similar in magnitude to the wavelength of light in the GaAs. Good angular randomization should also result in a strong suppression of the Fabry-Perot (FP) effects which occur as a result of the cavity formed by the sample surface and the backside mirror. This could be observed in both PL and photocurrent measurements. Within the linewidth of the PL signal, 3 periods of Fabry-Perot oscillations could be observed by dividing the spectrum by the average spectrum of all samples. In the average spectrum, the FP oscillations are eliminated, as the selected samples were taken from different positions on the wafer with slightly varying layer thicknesses. For the photocurrent samples, the relative deviation of the signal from an average curve fit to the photocurrent spectrum between the AlGaAs and GaAs bandgap was taken as a measure of the relative amplitude of the FP oscillations. The results of this evaluation, which are shown in Fig. 8, show a clear reduction of the oscillations for bead sizes between 100 and 300 nm for both types of samples. This maximum reduction is slightly shifted to smaller bead sizes compared to that of the maximum extraction efficiency.

Fig. 8: Amplitude of the Fabry-Perot oscillations for PL and photocurrent measurements, relative to the average signals. The error bars are the standard deviation of the various PL measurements. The solid line is just a guide to the eye.

3. LIGHT-EMITTING DIODES

Device fabrication

The optimum texturing conditions were applied to LED structures. The samples which were MBE grown consist of a 50 nm AlAs release layer for epitaxial lift-off, an n-doped AlGaAs cladding layer, a 120 nm

thick active n^- GaAs layer and a p-doped top cladding made of AlGaAs. In the top cladding, an $\text{Al}_{0.98}\text{Ga}_{0.02}\text{As}$ layer was included for lateral wet oxidation⁷ to provide current confinement⁸.

The initial processing step defined mesas of $50 \times 75 \mu\text{m}^2$ which were etched into the top AlGaAs cladding through the $\text{Al}_{0.98}\text{Ga}_{0.02}\text{As}$ layer in a manner such that it was close to the active layer but not completely through the cladding layer. The remaining p-AlGaAs layer is thus depleted avoiding current spreading and confining the current in the device¹. Then, the $\text{Al}_{0.98}\text{Ga}_{0.02}\text{As}$ layer was oxidized in wet N_2 at 400°C for 10 min., resulting in a lateral oxidation depth of $15 \mu\text{m}$. Small ohmic top contacts were applied on top of the mesa, above the oxidized area. In this way, the generation of light below the top contact could be drastically reduced. In a final processing step, mesas were etched to the bottom cladding and ohmic bottom-contacts were applied. The distance between the device mesa and the bottom contact mesa was kept large enough to avoid surface recombination in the active GaAs layer.

After this, the samples were cleaved into several test pieces. Each of them received systematically varying levels of surface texturing, as described in section 2. Finally, epitaxial lift-off was performed and a $\text{SiO}_2/\text{Ni}/\text{Au}$ back mirror was evaporated. In order to study the influence of the reflectivity of the mirror, one sample following epitaxial lift-off, was Van-der-Waals bonded to a gold mirror evaporated on a silicon wafer and coated with polyimide.

Experimental results

The LEDs emit at a wavelength of 875 nm with a linewidth of 30 nm (FWHM). The angular distribution of all LEDs, non-textured as well as textured, is lambertian. Fig. 9 shows the increase of the light output of the textured devices with an evaporated backside mirror compared to that of the “ordinary” LED, i.e. before texturing and epitaxial lift-off. A factor of 6 increase in the efficiency could be obtained for bead sizes between 200 and 400 nm and for an etching depth of 160 nm. The optimum bead size is in perfect agreement with the results of the PL study.

Fig. 9: Enhancement of light output of an LED structure obtained by application of a back mirror and surface roughening. The etching depth for all samples is 160 nm. Please note that the range of bead sizes investigated is smaller than in our PL studies, when comparing this figure to Fig. 6.

In Fig. 10 the external quantum efficiency (corrected for the respective emission wavelength) of various devices is shown. The “ordinary” device without the mirror and roughening exhibits a maximum external quantum efficiency of 1.7%, which corresponds to 2.4×10^{-2} mW/mA. This efficiency is obtained between 2 and 8 mA device bias currents. The decrease at lower currents is caused by nonradiative recombination, while the decrease for higher currents is due to heating/thermal effects.

By application of an evaporated back mirror the efficiency could be increased by a factor of 2, while surface texturing without the mirror results in no improvement of the efficiency. As already shown in Fig. 9, roughening and application of a back mirror results in a total increase of a factor of 6, at least for low currents. For these samples with an evaporated back mirror, the maximum efficiency is reached for a relatively low device current. This can be explained by the low thermal conductivity of the optical glue with which the samples were fixed onto their holder, which implies that the heating effects occur already for smaller device currents. Due to this limitation, the maximum quantum efficiency obtained with these devices was 6% which corresponds to 8.5 mW light output per 100 mA device current.

Fig. 10: External quantum efficiency for variously treated LED structures versus device current. All roughened LEDs were textured using 200 nm beads and an etching depth of 160 nm. The maximum quantum efficiency corresponds to a light output of 0.14 mW/mA.

The experiment with the Van-der-Waals bonded polyimide/gold mirror was carried out on a device from a different growth with an almost identical structure. As the reference device without the mirror had a maximum efficiency similar to that of the first growth, the results of the textured samples with a back mirror from the two different growths could be compared. These samples show a maximum quantum efficiency close to 10% over a wide current range from 1 to 10 mA. This corresponds to an efficiency of 14×10^{-2} mW/mA. The thermal behavior of these samples is comparable to the devices before epitaxial lift-off, which implies that this kind of mirror provides good heat dissipation. An efficiency of more than 1%

was obtained for the whole current range between 10 μ A and 10 mA, where the total output power ranged from 0.2 μ W to 1.4 mW.

4. CONCLUSIONS

We have presented a systematic study of the various parameters of surface texturing and its implications with specific reference to its application in light-emitting devices. These parameters were extracted from photoluminescence structures resulting in optimum sizes of the spheres used in the natural lithography of 200 to 300 nm. The texturing resulting from the etch process performed with the spheres acting as a mask should extend to a minimum depth of 160 nm. We expect a larger etching depth to result in a further improvement of the light output. In order to ensure that the enhancement of the PL signal is mainly caused by the improvement of the extraction of light, the study has been accompanied by experiments on detector structures. These experiments proved that, for the conditions we used, the PL enhancement is mainly caused by an improved extraction of light by surface texturing and the application of a back mirror. In addition, they also showed that surface texturing can increase the response of photodetectors considerably.

The application of the optimized parameters on real LED structures resulted in an external quantum efficiency of 10% for a 50 μ m by 75 μ m device. The study also showed that a small variation in the optimized parameters for surface texturing is not critical, which makes the concept of surface texturing very promising for the fabrication of light-emitting devices.

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