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Enhanced light extraction from InGaN/GaN quantum wells with silver gratings

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We demonstrate that an extraction enhancement by a factor of 2.8 can be obtained for a GaN quantum well structure using metallic nanostructures, compared to a flat semiconductor. The InGaN/GaN quantum well is inserted into a dielectric waveguide, naturally formed in the structure, and a silver grating is deposited on the surface and covered with a polymer film. The polymer layer greatly improves the extraction compared to a single metallic grating. The comparison of the experiments with simulations gives strong indications on the key role of weakly guided modes in the polymer layer diffracted by the grating. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4794066]

Indium gallium nitride (InGaN) quantum wells (QW) are widely used for optoelectronic devices, and are nowadays the key to energy-efficient lighting and display components. Bright efficient InGaN QW light emitting diodes (LEDs) can indeed be obtained, and with a large range of wavelengths (blue to green).[1] However, this inorganic, thus, very reliable material, has a very high refractive index (2.4 at 500 nm wavelength), resulting in a limited light extraction for conventional designs.

Various methods have been proposed to increase the efficiency of InGaN diodes.[2–5] One of them consists in using metallic structures to enhance the radiative rate thanks to the surface plasmons they support.[6,7] These plasmon modes have a high local density of states, and consequently catch the main part of the emission, which can then be coupled to radiative light with a corrugation (either periodic[6] or not[8,9]). Luminescence enhancements of up to 15 have been demonstrated with these processes. Nevertheless, extraction enhancement based on surface plasmons is hardly compatible with an electrical excitation. Indeed, the efficient coupling of the emission to the surface plasmon requires a close proximity between the emitters and the metal. The metal also creates a depletion region in the semiconductor, making the electrical excitation difficult.[10] An efficient alternative is based on etching periodically the GaN waveguide, which brings back into the light cone the emission trapped into the semiconductor structure. Even if the radiative emission rate of the QW remains constant, a large increase in the light extraction has been successfully demonstrated by several researchers using this method.[11,12] In addition to enhancing the efficiency, this method improves the emission directivity. However, this technique requires a deep etching of the semiconductor, which is especially challenging for GaN and requires expensive lithography.

We propose in this paper a method to enhance the light extraction of GaN QW using periodic structures, but avoiding a complicated etching of the structure. For this purpose, we use a metal not because it supports surface plasmons, but because of its high index contrast with the semiconductor. InGaN/GaN QWs are usually grown on a sapphire substrate, which means that they are naturally inserted in a waveguide structure delimited by sapphire on one side and air on the other side. Thus, a predominant part of the light is emitted into guided modes.[13] Our method consists in diffracting efficiently these waveguided modes trapped in the structure using metallic gratings deposited on the InGaN/GaN QW structure. However, even if the index contrast between the metal and the semiconductor material is large, it is not sufficient to efficiently diffract the light emitted in the waveguide, as the mode intensity at the grating is very small. A polymer layer (PolyVinyl Alcohol, PVA) has thus been spin-coated on the metal, which creates waveguided modes in the PVA film. With these combined (technically simple) processes, an enhancement of the luminescence by a factor of 2.8 over the bare structure has been obtained. In addition to the extraction enhancement and the control of the light directivity, this method has technological advantages: it can be simply applied to a wide variety of LED structures and is compatible with soft lithographic methods.

Samples, schematically presented in Figure 1, were grown on c-plane sapphire Al_2O_3 substrates by metal organic vapour phase epitaxy (MOVPE). The N, In, and GaN sources are, respectively, ammonia (NH_3), trimethilindium (TMIn), and trimethylgallium (TMGa). The 3 nm thick $In_{0.12}Ga_{0.88}N$ QW is grown on a 4 μm GaN buffer layer. It is then capped with a 20 nm GaN barrier. The roughness of the surface is very small, about 3 nm according to atomic force microscopy (AFM) results (not shown). The QW emits at 475 nm. Ag gratings have been defined by e-beam (EB) lithography on poly(methyl methacrylate) (PMMA) resist deposited on the

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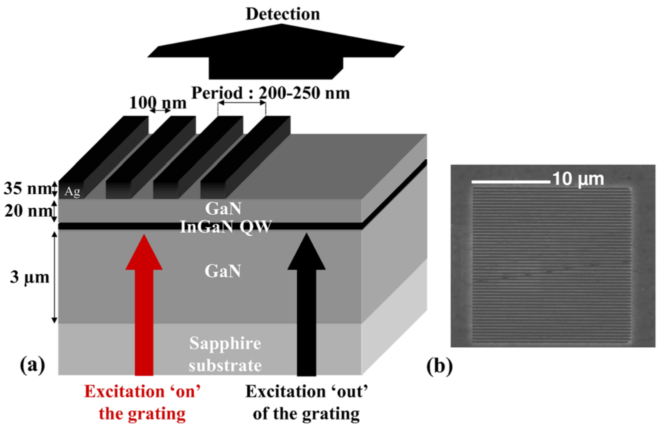

FIG. 1. (a) Schematic representation of the sample. (b) Transmission electron microscopy image of a silver $20 \times 20\,\mu m^2$ square.

InGaN/GaN heterostructures. A 35 nm thick silver film was evaporated and then removed by the lift-off technique, hence revealing silver gratings on the surface. Gratings with different periods were deposited, consisting of $20\,\mu$m squares spaced from each other by more than $100\,\mu$m. A particular care was devoted to the realization of the EB exposure, since it has been shown that even low energy electrons can destroy the InGaN QW when they are too close to the surface.[14,15]

The structure was excited by a 405 nm laser diode, focused through the substrate on the QW. The size of the laser spot on the sample was $5\,\mu$m, smaller than the size of one silver square ($20\,\mu$m). The emission of the sample was then collected from the top of the sample with a microscope objective (Numerical Aperture 0.7). The Fourier plane of the microscope objective was imaged on the entrance slit of a spectrometer coupled to a charge-coupled device (CCD) detector, allowing the determination of the angular dependence of the emission. Spectra integrating the total intensity emitted in the detection cone of the microscope can also be taken.

To quantify the luminescence enhancement obtained with the metallic grating, two QW luminescence spectra were recorded and compared: one when the laser pumping spot was focused under a periodic structure and another when it was focused outside the structure, i.e., under a bare surface (close to the studied structure). As the sample is excited through the substrate, we assume that the QW is excited in the same way whether the surface is covered by a grating or not.[6] The change in the extraction efficiency can be obtained by calculating the ratio between the maximal intensity under and outside the metallic grating. To clearly separate the effect of the material forming the grating and the covering layer, three different types of corrugations were investigated: a polymer grating, a metallic grating, and a metallic grating covered with PVA. The polymer grating has the same period as the metallic one and its height is about 100 nm. Figure 2 shows the luminescence spectra recorded under the grating structures and outside them, for the different configurations, each for a 200 nm period. In Figure 2(a), the luminescence recorded in transverse electronic (TE) polarization on the sample with a PMMA grating show no specific enhancement compared to the bare GaN structure. The ratio of enhancement in the case of a silver grating (Figure 2(b), TE) is about 1.2. To enhance the diffraction efficiency of the metallic grating, the metal could be physically buried in the guide but this would be complicated from a technological point of view. A simpler solution consists in depositing another high index layer on the metallic grating, which is approximately equivalent to burying the grating in the waveguide. For this purpose, a thick PVA layer was deposited on the whole surface of the sample. A very intense luminescence was then observed, the ratio to that of the bare structure reaching up to 2.8 (Figure 2(c), TE). This is comparable with an enhancement that could be expected from a perfectly diffusive surface.[19] Figure 2(d) shows the luminescence recorded under the silver grating with PVA, but in

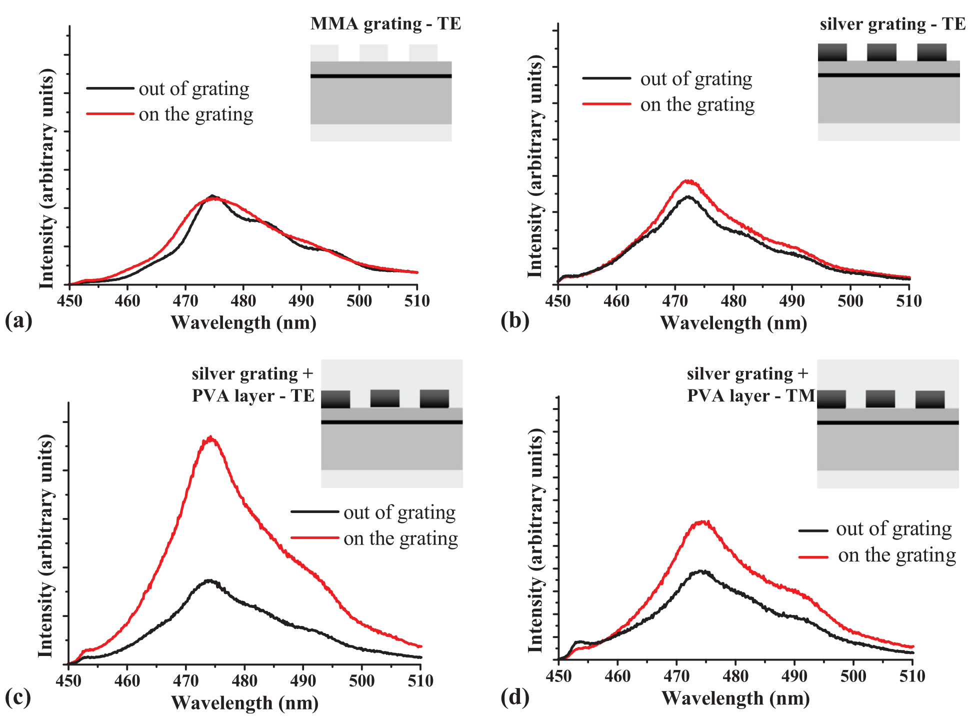

FIG. 2. Luminescence under the grating (red lines) and outside the grating (black lines): (a) PMMA grating in TE polarization (ratio about 1), (b) silver grating in TE (ratio about 1.2), (c) silver grating covered with a thick PVA layer in TE (ratio about 2.8), (d) PVA coated silver grating in TM (ratio about 1.6).

transverse magnetic (TM) polarization, where a smaller ratio was obtained: 1.6. This figure demonstrates first that a significant luminescence enhancement can be obtained with metallic gratings covered with PVA, and second that the enhancement is higher in the TE than in the TM polarization. We have also obtained similar results (a mean ratio of 2.2 for metallic gratings with PVA in TE, data not shown) for a periodicity of 250 nm, so it seems that the enhancement is not highly sensitive to small period modifications.

To investigate the origin of this luminescence enhancement, spatially and angle-resolved experiments were conducted on the sample with a silver grating of period 200 nm covered with a PVA layer. The Fourier-imaged reflectometry results were performed in TE polarization (Figure 3(a)). Three different contributions can be observed in the reflectometry image. Interference fringes in an arc shape repeating about every 12 nm are present, superposed on larger arcs every 25 nm. These patterns are caused by Fabry–Perot resonances in the different layers forming the structure: the GaN waveguide sandwiched between the sapphire substrate at the bottom and the metal at the top, and the PVA layer between the metal and the air. The maxima measured are consistent with the 4 μm thickness of the GaN layer and with the 2 μm thickness of the PVA layer. Furthermore, the larger arcs are not present when the PVA layer is not deposited on top of the structure (data not shown). In addition to these smooth variations in the reflectometry, two series of sharp lines are present in the image, with ascending and descending slopes. A similar pattern has been obtained for waveguides with etched gratings.[11] These lines are associated with the diffraction of the waveguided modes which are brought back into the light cone by the grating. To confirm this interpretation, the modes of the structure have been simulated and are presented in Figure 3(b). The modeling of the reflectometry measurements of the grated structure is performed by first using the transfer matrix method to solve the electric field profiles due to the incident light and the Green's functions of the planar structure with a 17 nm thick metal layer.[17] This 17 nm thickness was chosen to take into account the 35-nm thick half-filled silver film. Changing this thickness would not affect the dispersion image that has been extracted from this simulation. A perturbative approach using the Green's functions to evaluate the scattered field was then used to account for the scattering by the metallic grating consisting of 100 periods of 200 nm.[18] Further details of the method will be published elsewhere.

To go further into the analysis of the waveguided modes, the electric field associated with the modes contributing to the features observed in Figure 3 at different positions of the dispersion image were calculated (Figure 4), and two types of guided modes can be separated. Only guided modes in GaN are scattered to the region in between the dashed lines drawn in Figure 3(b). An example of a GaN mode marked as trace 1 is shown in Figure 4. Above and below the dashed lines (traces 3 to 5), GaN guided modes coexist with modes guided in the PVA layer. These last modes only exist because of the metal between the PVA and the GaN. Indeed, without the metal, as the sapphire substrate has a greater index than PVA, field confinement would not be possible. The field calculated for the PVA modes also has a non-vanishing part in the GaN layers. It should be noted that the GaN modes appear with a very low contrast in the calculated dispersion images and cannot be seen in the experimental spectra.

Figure 5 shows the angle-resolved luminescence images. Figure 5(a) shows an uncovered QW sample, and no specific structuration of the luminescence can be seen, except for the interference fringes in an arc shape corresponding to the GaN layer. Apart from this Fabry–Perot pattern, the luminescence is homogeneous with the angle. On the other hand, Figure 5(b) recorded on the metallic grating covered with PVA shows a strong structuration of the luminescence with the angle. Bright sharp lines are present at the same position as the sharp lines in the reflectometry spectrum. It is very clear that high luminescence is obtained specifically on the bright

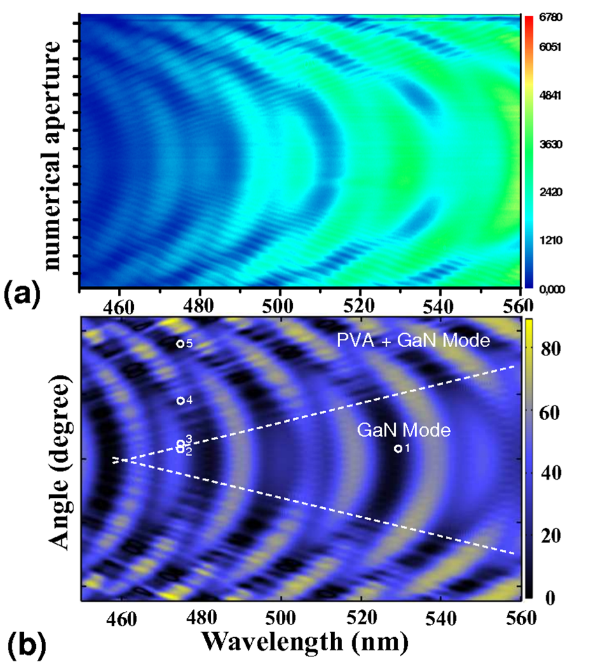

FIG. 3. Angle resolved reflectometry: (a) measurements on a structure with a silver grating and a PVA layer in TE polarization, (b) simulation of the same structure. The circles indicate the position where the fields are drawn in Figure 4. Dashed lines separate the area where only GaN modes are scattered from the area where PVA modes also contribute.

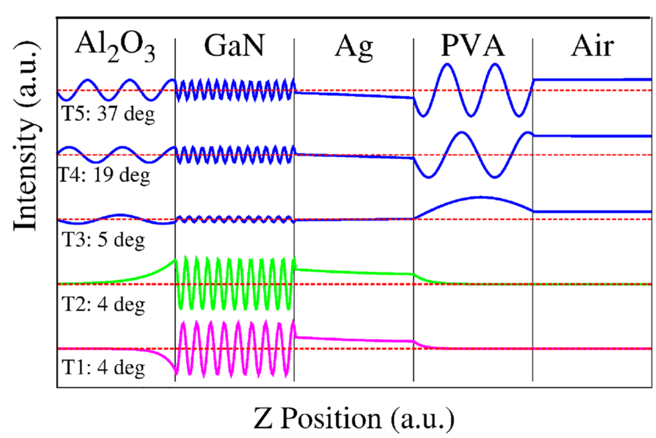

FIG. 4. Simulation of the electric field calculated for modes at the positions of the dispersion relation pointed in Figure 3(b). Trace 1: $\lambda = 529$ nm; traces 2 to 5: $\lambda = 475$ nm.

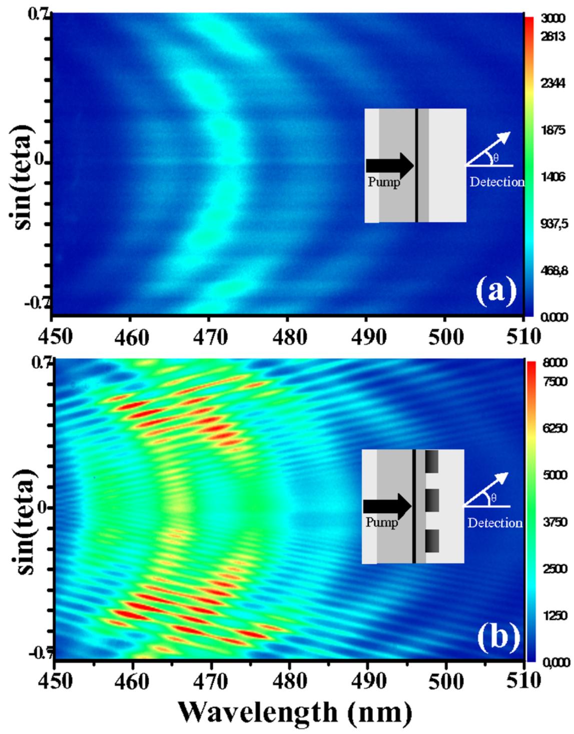

FIG. 5. (a) Experimental angle-resolved luminescence outside any grating structure (b) Experimental angle-resolved luminescence under a silver grating of period 200 nm covered with a thick PVA layer.

little lines and that a clear angle dependence appears. We can therefore conclude that strong luminescence enhancement finds its origin in the diffraction of the waveguided modes by the metallic grating. Moreover, the enhancement is only present in the region where the PVA modes are located (above and below the dashed lines in Figure 3(b)), confirming the key role of the PVA modes in the extraction enhancement.

The above discussion concerns TE polarization. In TM polarization, the luminescence enhancement is reduced: an enhancement by a factor of 1.6 was measured (Fig. 2(d)) for metal covered with PVA. A hypothesis to explain this reduction could be that there is a coupling of the TM emission to the surface plasmon modes, something which produces more losses than the guided modes. The coupling with the plasmons does not affect the TE emission, as plasmons only exist in TM. To confirm this interpretation and improve the TM extraction, the distance between the QW and the metal grating should be analysed. The shape of the metallic grating could also be modified, based on the work performed on etched photonic bandgaps in GaN.[16] A detailed analysis of the optimal metal structure (period, design) and position of the QW to prevent plasmon coupling is now under study but is outside the scope of this letter. Nevertheless, a metallic grating could not be replaced by a high index material corrugation to enhance the extraction with the leaky modes appearing in the PVA. Indeed, guided mode in the PVA cannot be obtained if deposited on a higher index layer. The metal layer is thus necessary to generate guided modes in the silver/PVA/air layers.

To summarize, we have shown experimentally that an efficient light extraction can be obtained with metal gratings on top of InGaN/GaN QW structures, covered by a polymer film. The diffraction is efficient thanks to the high index difference between the metal and the semiconductor. The PVA layer has a major role in the extraction enhancement, which can be related to the diffraction of the guided modes it supports. An enhancement by a factor of 2.8 has been obtained over the uncovered QW. As this technique only requires surface modification, it could be easily applied to existing diodes without complex etching. The method proposed is also compatible with soft lithography methods such as nanostamping, allowing large scale fabrication.

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