Optical gain in optically pumped cubic GaN at room temperature

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We investigate the transient surface emission and the optical gain of cubic GaN on GaAs(001) upon pulsed optical pumping at room temperature. The initial decay time of the transient surface emission drastically decreases with increasing excitation density, reaching a value as short as 20 ps at a fluence of 50 μJ cm^{-2}. This rapid decay suggests the presence of laterally amplified spontaneous emission. In fact, gain-stripe measurements of the edge emission reveal an optical gain exceeding 100 cm^{-1} at a fluence of 20 μJ cm^{-2}. © *1997 American Institute of Physics.*
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The demonstration of GaN-based light emitting devices[1] in the blue spectral region has resulted in worldwide efforts to realize GaN-based laser diodes. In fact, optical gain at cryogenic temperatures was reported decades ago for a single crystal needle of GaN.[2] Optical gain at room temperature has been observed for epitaxial GaN layers on different substrates[3–5] as well as for InGaN/GaN (Refs. 6 and 7) and GaN/AlGaN (Refs. 8–10) heterostructures. These structures were prepared on sapphire or SiC substrates by either metal-organic vapor phase epitaxy (MOVPE),[3–8] or molecular beam epitaxy (MBE).[9,10] Very recently, a pulsed injection laser that operates at room temperature and consists of an InGaN multiple-quantum-well structure fabricated on sapphire by MOVPE was demonstrated.[11]

All structures for which optical gain has been observed so far are based on the hexagonal modification of GaN. To the best of our knowledge, optical gain has not yet been reported for the cubic modification of GaN, which has been much less explored compared to the hexagonal modification because of its notoriously inferior quality. The cubic modification offers, however, distinct advantages over hexagonal in that it has a band gap closer to the blue spectral range and cleavage planes compatible to those of technologically attractive substrates such as GaAs. In this letter, we report the observation of optical gain in a cubic GaN layer at room temperature. The net gain exceeds 100 cm^{-1} at a fluence of 20 μJ cm^{-2}.

The sample investigated was grown by solid-source MBE on GaAs(001) employing a dc plasma discharge source for dissociating molecular N_2 into activated nitrogen species. Further details of the growth and characterization of cubic GaN may be found in Refs. 12–14. The sample studied here is predominantly cubic as revealed by x-ray diffraction and Raman spectroscopy, and its surface is featureless under an optical microscope.

The emission spectra were recorded using 150 fs pulses from a frequency-doubled Ti:sapphire laser at a photon energy of 3.36 eV with a repetition rate of 76 MHz. The excitation power was reduced by neutral-density filters and controlled by a powermeter. For recording the surface emission, the sample was excited in backscattering geometry with the laser spot focused to a spot of $\simeq$100 μm in diameter. To record the edge emission, the excitation spot was focused onto a $l\times 50$ μm^2 stripe on top of the surface adjacent to a cleaved edge, where l denotes the excitation length. To measure the optical gain we used the method pioneered by Shaklee *et al.*,[15] in which l is varied with an adjustable slit. The light emitted was collected by a lense, dispersed by a 22 cm monochromator (600 lines/mm grating), and focused onto the photocathode of a streak tube that allowed a time resolution down to 2 ps. The streak images were recorded by a cooled charge coupled device (CCD)-array. The spectral resolution of the setup was about 3 meV. All measurements were performed at room temperature.

Figure 1 shows surface emission transients for different excitation power densities. The decay is monoexponential and rapid ($\simeq$10 ps) at low fluences. The decay time increases to values of $\simeq$1 ns with increasing fluence. At the highest fluence used, the initial decay accelerates and reaches a value as short as 30 ps. The solid lines in Fig. 1 originate from simulations based on a recently published model,[16] that considers parallel carrier recombination by band-to-band and defect-mediated transitions. Within this model, the rapid decay at low fluences is understood as being dominated by defect-mediated recombination. The decay decelerates once

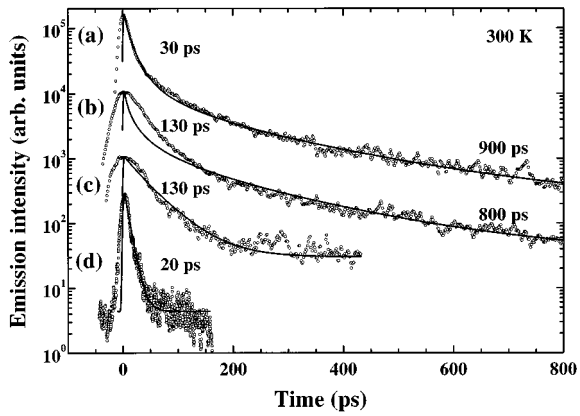

FIG. 1. Transients of the surface emission (symbols) in backscattering geometry at 300 K. The excitation fluences are 12, 3, 0.1, and 0.01 μJ cm^{-2} for transients (a), (b), (c), and (d), respectively. The traces are vertically shifted for clarity. The solid lines are fits of a recombination model. The major decay times are indicated.

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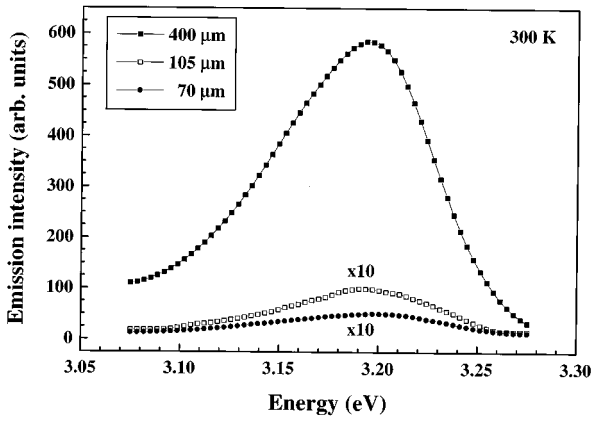

FIG. 2. Edge emission spectra for different excitation stripe lengths for a fluence of 20 μJ cm^{-2} at 300 K.

these defects are saturated at sufficiently high fluences, and thus recombination approaches the purely radiative regime. However, the rapid initial decay at high fluences cannot be understood within this model. It can be described only when an additional bimolecular term, which is characterized by a carrier-density dependent radiative recombination coefficient, is added to the equations. Although this phenomenological term is formally similar to that describing Auger recombination, we find the total (spectrally and temporally integrated) surface emission intensity to increase slightly superlinearly (exponent 1.2) with fluence. It thus seems more likely that the rapid initial decay originates from the onset of laterally amplified spontaneous emission.[17]

Figure 2 shows the edge emission spectra for several excitation lengths at a fixed power density of 20 μJ cm^{-2} at 300 K. The superlinear increase in intensity above an excitation length of $\simeq$100 μm is evident from these spectra. The spectral shape of the spectra, however, remains virtually unchanged. This finding is not unexpected, since the measurements are carried out in a single-pass configuration (i.e., there is no cavity).

Figure 3 shows the total edge emission intensity (symbols) as a function of the excitation stripe length for fluences of 0.6, 5, and 20 μJ cm^{-2} at room temperature. For the two

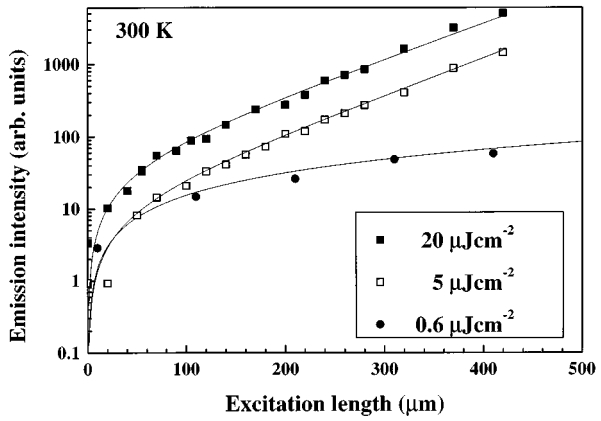

FIG. 3. Total edge emission vs excitation stripe length for different fluences at 300 K. The lines are fits of model calculations to deduce the respective gain values.

higher fluences, an exponential dependence of the edge emission intensity on excitation length is observed and reveals the presence of optical gain and thus of the onset of amplified spontaneous emission. In order to determine the gain coefficient g from the length dependence of the emission intensity I, we used the well-known formula

$$I=\frac{I_0}{g}[\exp(gl)-1], \tag{1}$$

where I_0 denotes the spontaneous emission rate per unit volume.[15] The calculated gain curves corresponding to the best fit to the data points are shown as solid lines in Fig. 3. The gain coefficient increases from 5 cm^{-1} at 0.6 μJ cm^{-2} to values between 110 and 120 for 5 and 20 μJ cm^{-2}. The latter values are comparable to the gain coefficients measured for hexagonal $Al_{0.1}Ga_{0.9}N$/GaN heterostructures.[8] Note that the carrier density created by an ultrashort optical pulse is proportional to the *fluence* of the pulse and not to its *intensity*. While the excitation intensities used here are significantly higher than in previous works, the fluences are not.

In summary, we have observed optical gain at room temperature in a cubic GaN layer with a thickness of about 1 μm. At a fluence of 20 μJ cm^{-2}, the gain coefficient is larger than 100 cm^{-1}. This observation raises the hope that cubic GaN may eventually reach maturity and become a viable material for applications. Future work in our laboratory will include the photopumping of cleaved cavities of cubic GaN on GaAs in an attempt to observe stimulated emission.

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