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# Luminescence and ESR Spectra of GaN:Si below and above Mott Transition

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Investigations of luminescence and ESR of silicon doped GaN layers are presented. The room temperature electron concentration in the investigated layers ranged from $1.7x10^{17}$ $cm^{-3}$ to $7x10^{18}$ $cm^{-3}$. The layer with the highest electron concentration has metallic conductivity. The ESR investigation revealed the presence of a characteristic asymmetric resonance whose intensity grows with increasing silicon impurity concentration. This resonance, corresponding to perpendicular g=1.985 and parallel g=1.983 has been observed in Si doped layers with electron concentration below the Mott transition. It seems that the ESR resonance is due to isolated Si donors. It has been found that the total PL emission increases with the silicon concentration, and is strongest in the layer with metallic conductivity. This indicates that silicon impurities eliminate non-radiative recombination centers or they create a new path of radiative recombination. The AFM and low temperature PL measurements indicate that strain relief via creation of pinholes may be responsible for the increase of radiative emission in GaN:Si epilayers.

## 1 Introduction.

The group III nitride technology has reached such a high level that the first blue cw laser operating at room temperature has been demonstrated [1]. For blue and UV emitters, the understanding of radiative and non-radiative recombination paths is of crucial importance. Electrical, ESR and PL measurements have been performed to understand the efficient radiative recombination channel in GaN:Si.

## 2 Experimental

GaN:Si was grown by the atmospheric pressure MOCVD technique, starting from TMG, $NH_3$ and $SiH_4$, and using (0001) sapphire substrates. The GaN buffer layer was grown at 560°C, followed by a Si doped epitaxial layer grown at 1060°C. All the layers have been grown under the same growth conditions, except for the monosilane flow rate, in order to get GaN:Si layers with net electron concentrations of $1.7x10^{17} – 7x10^{18}$ $cm^{-3}$. The thickness of all the epilayers was approximately 3.5µm. The thickness of the buffer layer, which is of critical importance, was chosen in such way that smooth layers with mirror-like surfaces without hexagonal hillocks were grown.

The Hall effect, electron spin resonance (ESR), atomic force microscope (AFM) and photoluminescence (PL) techniques were applied to characterize the physical properties of the GaN:Si layers. The electron concentration was determined using the van der Pauw configuration and PL was performed using the 325nm line of a He-Cd 10mW laser.

## 3 Experimental results and discussion.

Results of electrical measurements of four GaN:Si layers grown with various silicon concentrations are shown in Figure 1.

The layer designated MAR70, with electron concentration $1.7x10^{17}cm^{-3}$ and mobility $740cm^2/Vs$ at 300K, shows a clear freeze-out of electrons down to a concentration of $6x10^{15}$ $cm^{-3}$ at 40K. The increase of resistivity between 300K and 50K is about two orders of magnitude. The mobility of electrons reaches a very high value of 2000 $cm^2/Vs$ at 120K. The decrease of mobility below 100K is due to hopping conductivity which takes place in the buffer layer.

The layer designated MAR68, with electron concentration $6.9x10^{17}$ $cm^{-3}$ and mobility $530cm^2/Vs$ at 300K, shows an increase of resistivity by one order of magnitude during cooling down to 50K. The layer designated MAR77, with electron concentration $1.6 \times 10^{18}$ $cm^{-3}$ and mobility 280 $cm^2/Vs$ at 300K, seems to be close to the Mott transition. The increase of resistivity with decrease of temperature observed in this layer is due to the decrease of mobility. The layer designated MAR78, with electron concentration $7 \times 10^{18}$ $cm^{-3}$ and mobility 200 $cm^2/Vs$ at 300K, is clearly above the metal – insulator transition. Conductivity in this layer is metallic-like without any activation energy.

These results are in agreement with the ESR measurements, shown in Figure 3. The layer MAR70 shows an anisotropy of spin resonance (for perpendicular g=1.985 and parallel g=1.984), the same as was reported previously for the residual donor in undoped GaN [2]. An increase of the ESR signal is observed for the MAR68 layer, which seems to be connected with an increase of the isolated silicon donor concentration. In spite of the fact that the hyperfine interaction is not resolved, we think that the observed ESR signal is due to isolated silicon donors. However, in the MAR77 and MAR78 layers the ESR silicon donor signal becomes smeared out. It seems that it is due to interaction between silicon donors, which leads to formation of the donor band.

Room temperature PL is dominated by the free exciton emission band. It has been found that the total UV emission increases with increase of silicon concentration. The integral over the UV emission band plotted versus Si concentration is shown in Figure 4. An increase of silicon concentration by a factor of 50 leads to an increase of the total UV emission by one order of magnitude. Such an increase of the total UV emission with increase of Si concentration has been previously reported [3]. However, the mechanism of this process is still unexplained. This is indeed a surprising result, as one would expect that above the Mott transition the nonradiative Auger recombination should effectively compete with the band-to-band radiative emission, decreasing the total emission. The results indicate, that instead, the increase of silicon concentration closes existing non radiative channels or opens up a new path for the radiative recombination.

In our previous study of homoepitaxial growth [4] we found that a smooth layer with eventual pinholes is characteristic of gallium terminated growth. On the other hand the presence of hexagonal hillocks is characteristic of the terminated growth. It seems that in the heteroepitaxial growth on $Al_2O_3$ the thickness of the buffer layer may determine the nitrogen or gallium terminated growth. The GaN:Si layers grown in exactly the same conditions but with a different thickness of the buffer layer shows different types of morphology (smooth with pinholes and rough with hexagonal hillocks). Despite having similar electron concentrations, close to $8 \times 10^{17}$ $cm^{-3}$, these two layers have have different optical properties. The PL spectra of the layers are shown in Figure 4. The low temperature PL spectra are dominated by donor bound exciton (DBE) recombination. The DBE emission in the smooth layer (characteristic of gallium polarity) is observed at 3.485eV. This indicates that the layer is strongly strained. On the other hand the DBE emission in the rough layer with hexagonal hillocks (nitrogen polarity) is observed at 3.472eV. This energy corresponds exactly to the DBE emission in the unstrained homoepitaxial [4] layer, indicating that the layer grown with hexagonal hillocks is fully relaxed. The electron mobility at room temperature has been found to be 400$cm^2/Vs$ in the strained layer and a factor of two smaller in the relaxed layer.

AFM and low temperature PL studies have been performed to investigate the evolution of stress in the GaN:Si layers. The results of PL are shown in Figure 5. A single peak dominates the PL spectra of layers containing low concentrations of silicon. The temperature dependence of the dominant emission peak indicates the characteristic shift from the donor bound exciton (DBE) emission to free exciton emission at higher temperatures. Therefore we believe that it is due to DBE recombination. The DBE emission line is observed at 3.4855eV for the MAR70 layer. The energy of the DBE line indicates that the layer is strongly strained. An increase of silicon concentration to $6.9 \times 10^{17}$ $cm^{-3}$ shifts the DBE emission line to higher energy, 3.487eV. This indicates that an increase of silicon concentration leads to an increase of strain in the layer. A further increase of silicon concentration to $1.6 \times 10^{18}$ $cm^{-3}$ leads to a dramatic change in the DBE emission peaks. We believe that it is a manifestation of strain relief in the layer. There are now two peaks at 3.474eV and 3.483eV, which we think are still due to DBE emission from regions of low strain (the 3.474eV line), and relatively high strain (the 3.483eV line). A further increase of silicon concentration up to $7 \times 10^{18}$ $cm^{-3}$ leads to a shift of the DBE emission line to 3.472eV, the energy corresponding to the emission in the unstrained homoepitaxial layer. The AFM investigation indicates that the main mechanism of strain relief is connected with the formation of pinholes. The layers with electron concentration of $1.7 \times 10^{18}$ $cm^{-3}$ and $6.9 \times 10^{17}$ $cm^{-3}$, in which strain builds up, does not show any pinholes. However, the layer with electron concentration of $1.6 \times 10^{18}$ $cm^{-3}$ shows the presence of pinholes with a density equal to $1 \times 10^6$ $cm^{-2}$. Further increase of Si doping, corresponding to electron concentration $7 \times 10^{18}$ cm$^{-3}$, is connected with a pinhole density equal to $4.5 \times 10^{6}$ cm$^{2}$. The average diameter of a pinhole is above 3 µm which lead to 30%-40% of area of the layer being covered by pinholes. Therefore, it may be expected that in this case the strain is completely relieved. This conclusion is in agreement with previously reported results indicating that an increase of Si doping leads to relaxation of residual stress in epilayers [5].

Free excitons and excitons bound to Si donors will have about 15meV lower energy in the strain-free regions. Therefore, confinement of excitons will take place in the unstrained regions. Apparently the confinement of excitons leads to an increase of band-to-band radiative recombination. Increasing silicon concentration increases the strain-free regions and leads to the increase of exciton emission via confinement of the excitons.


## ACKNOWLEDGMENTS

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## FIGURES

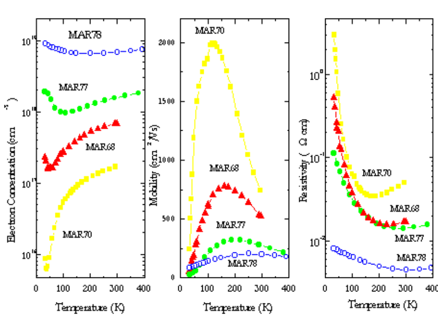


Figure 1. Temperature dependence of electron concentration, mobility and resistivity of four GaN:Si layers.

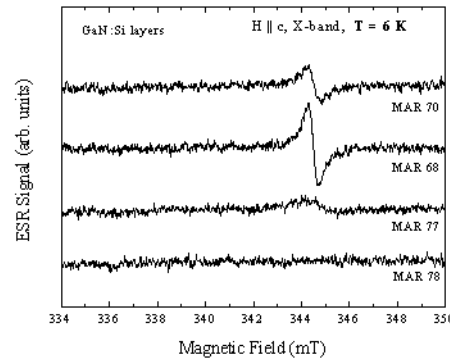


Figure 2. ESR spectra of GaN:Si layers.

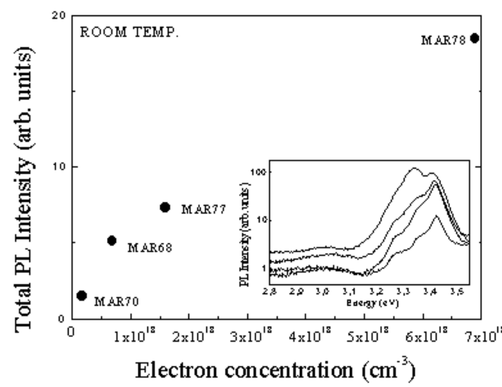


Figure 3. Integrals of UV photoluminescence emission at room temperature. The room temperature PL spectra of GaN:Si layers in the UV region are plotted in the insert.

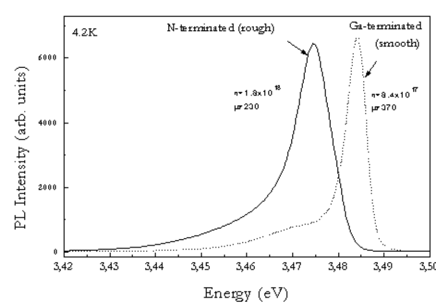


Figure 4. The low temperature photoluminescence spectra of two GaN:Si layers of different polarity grown in the same conditions. Solid line is the spectrum of relaxed layer, N-terminated (rough with hexagonal hillocks). Dashed line represent spectrum of strained layer, Ga-terminated (smooth with mirror like surface).

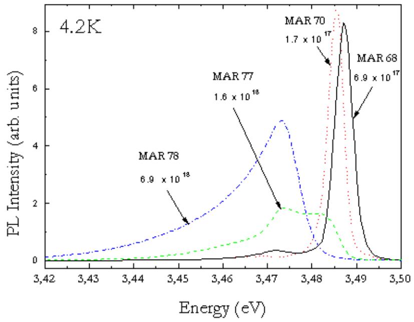


Figure 5. The low temperature photoluminescence spectra of GaN:Si layers. (The room temperature electron concentrations are given below the sample names.)



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