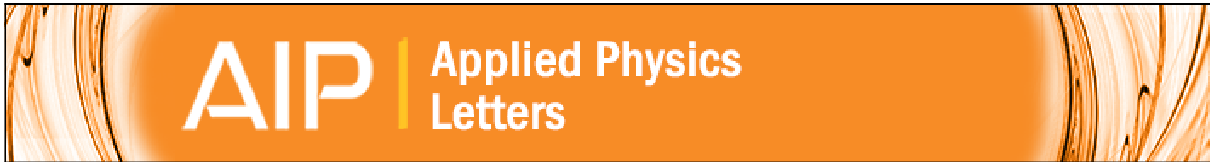

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Effect of annealing on the magnetic properties of Gd focused ion beam implanted GaN

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The authors have studied the effect of annealing on the magnetic and the structural properties of Gd focused ion beam implanted GaN samples. Molecular beam epitaxy grown GaN layers, which were implanted with 300 keV Gd^{3+} ions at room temperature at doses 2.4×10^{11} and 1.0×10^{15} cm^{-2}, are rapid thermally annealed in flowing N_2 gas up to 900 °C for 30 s. X-ray diffraction results indicate the presence of Ga and N interstitials in the implanted layers. Their densities are also found to reduce upon annealing. At the same time, magnetic measurements on these samples clearly show a reduction in the saturation magnetization as a result of the annealing for the lowest Gd incorporated sample, while in the highest Gd incorporated sample it does not change. These findings suggest that Gd might be inducing magnetic moment in Ga and/or N interstitials in giving rise to an effective colossal magnetic moment of Gd and the associated ferromagnetism observed in Gd:GaN. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2770762]

Recently, an extraordinary magnetic behavior has been observed in very dilute Gd doped GaN (Refs. 1 and 2) epitaxial layers. Even with a Gd concentration of less than 1×10^{16} cm^{-3}, the material was found to exhibit ferromagnetism.[2] Moreover, the average value of the effective magnetic moment per Gd atom was found to be as high as $4000\mu_B$, as compared to its atomic moment of $8\mu_B$. Similar effects have been reported in weakly Eu doped GaN (Ref. 3) as well. These findings, in one hand, have created exciting opportunities for application in spintronics and optoelectronics,[4] on the other hand, have generated several fundamental questions. In order to explain such a colossal magnetic moment, one has to consider a Gd induced long range spin polarization either of the GaN matrix atoms (Ga or/and N) or of a certain type of defects residing in the GaN matrix.[2] However, the microscopic origin of such a long range spin polarization is not clear yet. Very recently, Gd implanted GaN (Refs. 5 and 6) and AlN layers[5] are also found to exhibit a very similar magnetic behavior. Interestingly enough, the magnetic moment per Gd atom in these samples is found to be even larger as compared to that found in epitaxially grown layers for a given Gd concentration.[6] Since the implanted samples are expected to have a larger density of defects, this finding suggests that, defects play an important role in giving rise to the effect.[6] Annealing at high temperatures is expected to reduce the overall concentration of defects in the implanted layers. It would be thus interesting to study the change in the magnetic properties of these layers because of annealing. Such a study will be helpful not only to reach a firm conclusion over the role of defects in giving rise to this effect but also could give us a unique control to tailor the magnetic property through defect engineering which would be highly interesting as far as spintronic application is concerned.

In this letter, we investigate the effect of annealing on the structural and the magnetic properties of the GaN films implanted with Gd by focused ion beam (FIB) implantation technique at different doses. It has to be noted that these are the same set of samples, whose magnetic and the structural properties have been investigated in Ref. 6. Our study suggests that Ga and/or N interstitials might be responsible in giving rise to the effects of colossal magnetic moment of Gd and associated ferromagnetism observed in Gd:GaN.

Pieces of a 600 nm thick GaN layer, grown directly on 6*H*-SiC(0001) substrate by NH_3 molecular beam epitaxy, were implanted at room temperature with 300 keV Gd^{3+} ions at doses 2.4×10^{11} (sample A-1) and 1.0×10^{15} cm^{-2} (sample A-3), using a FIB implanter. These doses correspond to an average Gd concentration of 2.43×10^{16} and 1×10^{20} cm^{-3}, respectively. More details about the implantation could be found in Ref. 6. Implanted samples are rapid thermally annealed in flowing N_2 gas in two steps; at 800 °C for 30 s and then at 900 °C for 30 s. Before and after each annealing step, all the necessary structural and the magnetic characterizations are performed.

The structural properties of the layers were investigated by x-ray diffraction (XRD) using a Philips X'pert Pro™ diffractometer with Cu $K_{\alpha 1}$ radiation. Surface morphology of the films was examined by atomic force microscopy (AFM). Magnetization measurements from 4 up to 300 K were performed in a Quantum Design physical property measurement setup using the vibrating sample magnetometer head. The magnetization loops were recorded at various temperatures for magnetic fields between ±50 kOe with the field applied

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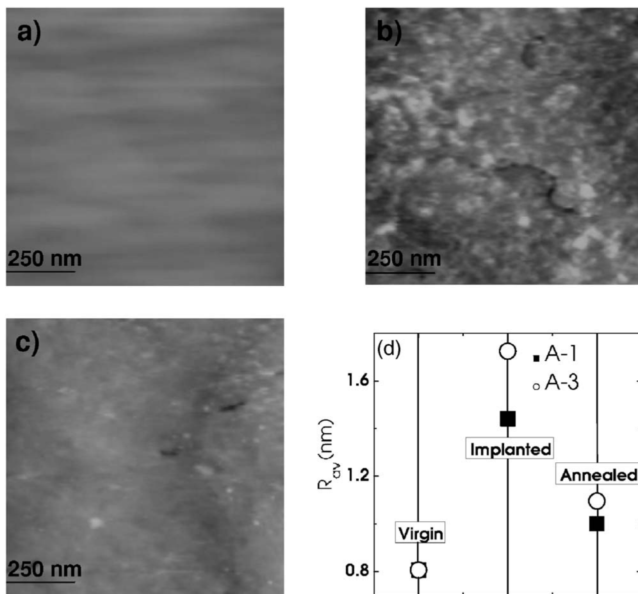

FIG. 1. AFM surface images of the GaN samples (a) before (b) after the Gd implantation at the highest dose, and (c) after the annealing at 900 °C for 30 s, (d) R_{av} as a function of the annealing and implantation states for both the samples.

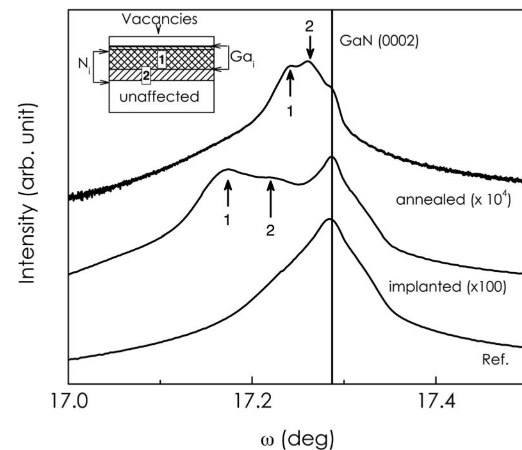

FIG. 2. X-ray diffraction ω-2θ symmetric scans taken on the reference sample (A-0) and sample A-3 before and after the annealing at 800 °C for 30 s. The inset is schematically showing the distribution of the vacancies and interstitials in the implanted layer.

parallel to the sample surface, i.e., perpendicular to the c axis. Field cooled (FC) and zero field cooled (ZFC) magnetizations are measured following the procedure described in Ref. 2. All data presented here were corrected for the diamagnetic background of the substrate.

Figures 1(a)–1(c) compare the AFM surface images taken on the GaN sample before and after the highest dose of Gd implantation and after the postimplantation annealing. It is clear from these panels that such a high dose of implantation results in an extensive damage on the surface, which, however, recovers by some extent upon annealing. Several $1\times 1\ \mu m^2$ AFM scans are taken on different parts of the sample. In Fig. 1(d), the overall roughness R_{av}, defined as the average of the individual rms roughnesses R_{rms} measured from these scans, is plotted as a function of the annealing and implantation states. Clearly, for both the samples, the surface roughness increases due to the implantation and reduces upon annealing.

Figure 2 compares the x-ray high-resolution triple-axis ω-2θ symmetric scans taken on the reference sample (A-0) and the sample A-3 before and after the annealing. Both for the annealed and unannealed cases, the GaN (0002) reflection appears almost at the same position and has a very similar width as that of the reference sample suggesting that this peak must be associated with the bottom part of the GaN layer which remains to be unaffected by the implantation. In addition to that, a clear broad feature appears at the left side of the GaN (0002) reflection both before and after the annealing. There are, in fact, reports of a satellite peak appearing at an angle slightly lower than that of the GaN (0002) reflection in XRD scans taken on GaN layers implanted with certain rare earth (RE) element[7] and few other non-RE elements[8] at energies and doses comparable to what have been used in this case. This feature was attributed to the expansion of the GaN lattice due to the formation of Ga and N interstitials by the ion bombardment. When examined closely, the feature is found to be actually composed of two peaks which are indicated by arrows 1 and 2. Their positions shift to the higher angles upon annealing which makes it unlikely that these features are stemming from any secondary phase.

Since Gd has a large atomic radius, its implantation into GaN is expected to results in a high density of Ga and N vacancies and interstitials. While the vacancies are likely to stay closer to the surface, the interstitials are supposed to be in excess at the end of range of the implanted ions.[9,10] Moreover, according to the theoretical work of Christel and Gibbons,[9] it is expected that the distribution profile for the N interstitials (N_i) will be displaced to a greater depth with respect to that of the Ga interstitials (Ga_i). It is plausible that the two peaks observed in this case are stemming from the different parts of the interstitial dominated zone; peak 1 is originating from the region which is dominated by both Ga_i and N_i, while the origin of peak 2 is the tail section of the N_i distribution profile, as shown in the inset of the figure. Since, region 1 is expected to be wider and more enriched with interstitials than region 2, peak 1 is likely to be sharper, stronger, and positioned at a lower angle than peak 2, which is consistent with the observation of Fig. 2 for the unannealed sample. Movement of these peaks to the higher angles after the annealing clearly suggests that the density of interstitials decreases due to annealing. Their relative intensities with respect to that of the GaN (0002) reflection (stemming from the bottom part of the GaN layer) increase upon annealing while, their respective width decreases, which indicates an expansion of both the zones as a result of the annealing. This suggests that the interstitials might be diffusing into the bottom part of the GaN layer which was hitherto been unaffected by the implantation. Note that the position, width and the relative intensity of these peaks do not change further upon annealing at 900 °C for 30 s suggesting that their densities might have reached a saturation already after the first annealing step. It is worth to be noted that in sample A-1, no satellite peak could be resolved from the GaN (0002) reflection. The reason might be an extremely low dose of implantation used in this case. However, full width at half maximum (FWHM) of the GaN (0002) reflection in this sample is found to be 122 arc sec which is more than its value of 79 arc sec found prior to the implantation. FWHM of the peak decreases to 97 arc sec upon annealing which suggests that the density of defects decreases due to annealing. One

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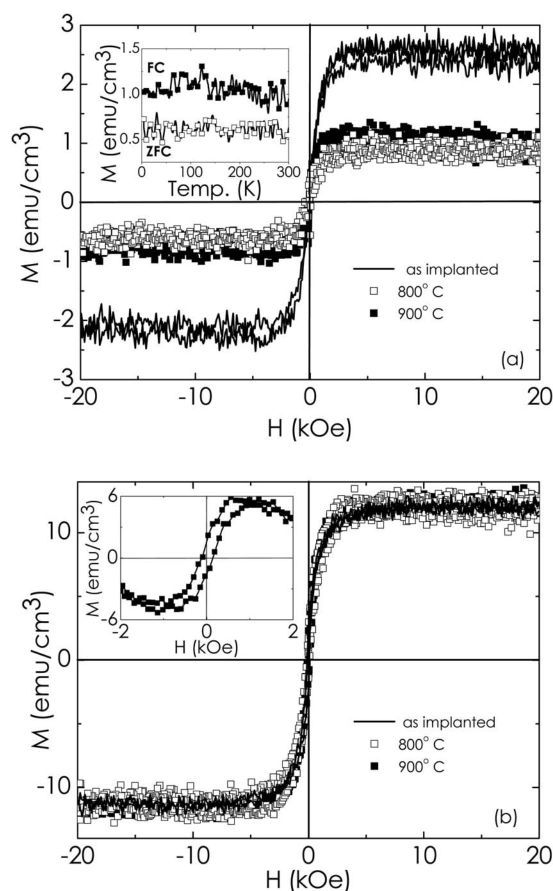

FIG. 3. Magnetization loops obtained at 300 K for (a) samples A-1 and (b) A-3 before and after the annealing at different temperatures. The inset of (a) shows the temperature dependence of the FC and ZFC magnetization measured at a magnetic field of 100 Oe for samples A-1 after the final step of annealing. The inset of (b) shows the magnetization loop at low fields obtained at 300 K for sample A-3 after the final annealing step before subtracting the substrate contribution.

should expect a very similar role of annealing on the diffusion mechanism of the interstitials, as has been observed in the case of sample A-3.

Figure 3 compares the field dependence of the magnetization measured at 300 K for sample A-1 [Fig. 3(a)] and sample A-3 [Fig. 3(b)] before and after each of the two annealing steps. In all cases, the magnetization exhibits a clear saturation at high fields. While a clear hysteresis could be seen in sample A-3 even after the annealing, as shown in the inset of Fig. 3(b), in the case of sample A-1 it is difficult to recognize. However, as shown in the inset of Fig. 3(a) a distinct separation could be seen between the temperature dependence of the FC and ZFC magnetizations for sample A-1 after the annealing which indicates a hysteretic behavior even at room temperature. In the case of sample A-1, the saturation magnetization (M_s) reduces by a factor of about 2 after the first annealing step. The second step of annealing does not change it any further, while in the case of sample A-3, M_s remains to be unaffected by the annealing. Magnetization loops taken at other temperatures in these samples also show a very similar dependence of M_s on the annealing.

The observations of these figures suggest that the decrease of M_s in case of sample A-1 must be associated with the reduction of the defects due to annealing. The results of the XRD characterization (Fig. 2) further indicate the presence of Ga and N interstitials in the implanted layers. The study also reveals a reduction of the density of these defects upon annealing. Since, it is well known that these interstitials are accumulated at the end of range of the implanted ions, Gd ions are expected to be surrounded mostly by interstitials. It is plausible that Gd induces magnetic moment in Ga and/or N interstitials in order to give rise to the effect of colossal magnetic moment. According to the sphere of influence model described in Ref. 2, the saturation magnetization contains two contributions. The contribution due to the Gd ions and the contribution from the defects which are magnetized by the influence of Gd. In sample A-1, $N_{Gd}=2.43 \times 10^{16}$ cm^{-3}, which is such that according to our previous observation from the N_{Gd} dependence of M_s, it should belong to regime II where the latter contribution is expected to dominate over the former.[2] On the other hand, sample A-3 with $N_{Gd}=1\times 10^{20}$ cm^{-3} should fall into regime III, where the contribution due to the Gd ions is supposed to dominate over the second. It is also consistent with the fact that p_{eff} is found to be $11\mu_B$ in sample A-3, which is very close to the Gd bare atomic moment of $8\mu_B$. This explains why in the case of sample A-1, M_s is found to decrease after annealing while in sample A-3 it does not change.

In conclusion, our study indicates that the colossal magnetic moment of Gd found in Gd implanted GaN is a result of a long range spin polarization of Ga and/or N interstitials generated due to Gd bombardment. In order to explain the same effect observed in epitaxial Gd:GaN layers, the presence of a large density (as high as 10^{19} cm^{-3}) of these defects in the layer is necessary. The theory, however, predicts[11,12] that these self-interstitials are quite unlikely to occur in GaN when it is grown at thermal equilibrium. In all cases, where the colossal magnetic moment of Gd has been reported so far, the Gd:GaN layers were grown using MBE. Since, in MBE, growth is carried out away from the equilibrium (low temperature, low pressure), formation of these defects cannot be ruled out. Another possibility, that Gd, with its large atomic size, is playing a catalytic role in the formation of these defects during growth, cannot be even excluded.

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[1] J. K. Hite, R. M. Frazier, R. Davies, G. T. Thaler, C. R. Abernathy, and S. J. Pearton, Appl. Phys. Lett. **89**, 092119 (2006).

[2] S. Dhar, O. Brandt, M. Ramsteiner, V. F. Sapega, and K. H. Ploog, Phys. Rev. Lett. **94**, 037205 (2005); S. Dhar, L. Pérez, O. Brandt, A. Trampert, K. H. Ploog, J. Keller, and B. Beschoten, Phys. Rev. B **72**, 245203 (2005).

[3] J. Hite, G. T. Thaler, R. Khanna, C. R. Abernathy, S. J. Pearton, J. H. Park, A. J. Steckl, and J. M. Zavada, Appl. Phys. Lett. **89**, 132119 (2006).

[4] S. Dhar, *Proceedings of the Department of Atomic Energy Solid State Physics Symposium*, Bhopal, India, 26–30 December 2006 (Manohar Chandnani, Mumbai, India, 2006), Vol. 51, pp. 25–28.

[5] S. Y. Han, J. Hite, G. T. Thaler, R. M. Frazier, C. R. Abernathy, S. J. Pearton, H. K. Choi, W. O. Lee, Y. D. Park, J. M. Zavada, and R. Gwilliam, Appl. Phys. Lett. **88**, 042102 (2006).

[6] S. Dhar, T. Kammermeir, A. Ney, L. Perez, K. H. Ploog, A. Melnikov, and A. D. Wieck, Appl. Phys. Lett. **89**, 062503 (2006).

[7] Bert Pipeleers, Susan M. Hogg, and Andre Vantomme, J. Appl. Phys. **98**, 123504 (2005).

[8] C. Liu, B. Mensching, M. Zeitler, K. Volz, and B. Raushenbach, Phys. Rev. B **57**, 2530 (1998).

[9] L. A. Christel and J. F. Gibbons, J. Appl. Phys. **52**, 5050 (1981).

[10] S. O. Kucheyev, J. S. Williams, C. Jagadish, J. Zou, and G. Li, Phys. Rev. B **62**, 7510 (2000).

[11] J. Neugebauer and C. J. Van de Walle, Phys. Rev. B **50**, 8067 (1994).

[12] P. Boguslawski, E. L. Briggs, and J. Bernholk, Phys. Rev. B **51**, 17255 (1995).

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