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Investigation of defects in Gd doped GaN using thermally stimulated current spectroscopy

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ABSTRACT

Defects in Gd-doped GaN layers are investigated using thermally stimulated current spectroscopy (TSC). The line-shape function, which commonly considers a delta function like density of state for the trap levels, is modified to take into account broad trap distributions. This function is used to fit the spectra which show two broad TSC features in all Gd-doped samples. The feature associated with the trap distribution peaking at about 45 meV from the band edge (TSC-1) can be attributed to unintentional oxygen donors, while the second feature for which the distribution peaks at ≈ 130 meV (TSC-2) from the band edge is assigned to the defects resulting due to Gd incorporation as the intensity of this feature increases with the Gd concentration. However, only a portion of the TSC-2 band, which is estimated to have an activation energy of 240 meV, is substantially reduced upon annealing at 800 °C. The annealing also results in a complete suppression of the magnetization. This implies that the observed magnetization is likely to be resulting from the defects associated with this portion of TSC-2 band. Furthermore, the energetic position of this portion of TSC-2 band (240 meV) matches very well with the activation energy (≈ 200 meV) for the low temperature PL peak at 3.05 eV, which has already been attributed to the defects responsible for the observed magnetic behavior. This suggests that both the TSC and the PL features are resulting from the same defect type and therefore, could provide a vital clue in the search for the exact nature of the defect responsible.

1. Introduction

Observation of certain intriguing magnetic properties in weakly Gd doped GaN layers has created tremendous excitements. Ferromagnetism far above room temperature for Gd concentration as low as $\approx 10^{15}$ cm^{-3} and several orders of magnitude enhancement of the effective magnetic moment per Gd ion as compared to its nominal magnetic moment of $7\mu_B$ are the hallmarks of the effect [1–4]. Similar properties have been reported in other rare-earth (RE) incorporated GaN such as Eu-doped GaN [5], Tm-doped (Al,Ga)N [6] as well as Gd- [7], Ce- [8], Er-implanted [9] GaN layers and in other RE incorporated semiconductors such as Gd-implanted AlN [10] and Gd-doped ZnO [11]. Observation of much higher saturation magnetization in implanted samples as compared to the samples doped in situ suggests that defects are the likely cause for the effect [7]. This conclusion is further supported by the fact that the saturation magnetization in implanted GaN:Gd layers is found to decrease upon annealing [12]. Furthermore, a large density of defects is reported to be generated during Gd incorporation in GaN:Gd layers doped in situ [13,3]. However, the identification of the defects responsible for the magnetic behavior in this material is still not conclusive. Different theoretical studies have attributed this magnetic property to different types of defects, such as Ga-vacancy [14,15,16], N-interstitials, O-interstitials [17] and vacancy complexes [18]. Recently, our study of low temperature photoluminescence (PL) spectroscopy on GaN:Gd layers doped in situ reveals that even though a variety of defects are formed in GaN layer during the Gd incorporation, only one defect type, which results in a strong luminescence feature at 3.05 eV, is responsible for the magnetic properties observed in this system as the intensity of this peak decreases upon annealing and at the same time, the saturation magnetization drops [19]. Investigation of defects in this material using standard electrical techniques, such as deep level transient spectroscopy (DLTS) and thermally stimulated current spectroscopy (TSC), can further shed light on the nature of the defects responsible for this effect. However, DLTS technique is useful only for conductive samples, while TSC can be carried out for high-resistivity samples [20]. Since, our Gd doped samples are found to be highly resistive, TSC is the only option left for such a study.

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In this letter, we explore the formation of defects in GaN:Gd epitaxial films using TSC. A band of trap levels is indeed observed in these layers, whose density is found to increase with the Gd concentration implying that they are resulting due to Gd incorporation. However, upon annealing at 800 °C, only a portion of the band is reduced substantially and at the same time, magnetization has been found to be suppressed completely. This finding points towards the defects associated with this portion of the TSC band to be the likely cause for magnetic behavior found in the system. Moreover, the energetic position of these traps is estimated to be very similar to the activation energy associated with the PL peak at 3.05 eV, which has already been attributed to the defects responsible for the observed magnetic properties. This indicates that they are resulting from the same type of defects.

2. Experimental

Gd doped GaN layers were grown directly on 6 H-SiC(0001) substrates by NH_3 assisted molecular beam epitaxy (MBE) technique. Details about the growth, structural and the magnetic characterizations of these samples are given elsewhere [2,13,19]. Here we have investigated samples with Gd concentration N_{Gd} $\approx 7\times10^{15}$ cm^{-3} (Sample A), $\approx 1.6\times10^{16}$ cm^{-3} (Sample B), $\approx 2.5\times10^{17}$ cm^{-3} (Sample D) and an undoped reference sample (Sample R). Thermally stimulated current spectroscopy (TSC) was carried out on these samples at temperatures ranging from 20 to 300 K using a closed-cycle helium cryostat. Sample was first cooled to 20 K and then illuminated with white light from a Xenon lamp for a sufficiently long time such that all traps are filled by the photo excitation. Subsequently, light was switched off and the temperature was raised to 300 K at a fixed rate β. At the same time, current through the sample was measured as a function of temperature for an applied bias of 0.1 V. In these measurements, β was maintained at different values ranging from 0.1 to 0.5 K/s. For electrical contacts, 1×1 mm^2 Ti/Au contact pads separated by a distance of 4 mm were fabricated on the sample surface. Sample D was rapid thermally annealed in flowing N_2 gas at 800 °C. TSC and magnetization measurements were carried out on the same piece of sample D before and after the annealing. Magnetization measurements at 300 K were performed on these samples in a Quantum Design physical property measurement setup using a vibrating sample magnetometer head for magnetic fields between $\pm$50 kOe applied parallel to the sample surface. All Gd doped GaN samples exhibit ferromagnetism and colossal magnetic moment effect at room temperature. Magnetization is found to be fully suppressed upon annealing in sample D [19]. Photoluminescence (PL) was carried out at temperatures ranging from 10 to 300 K with a He–Cd laser source (325 nm) for the excitation and a 0.5 m monochromator attached with a Peltier cooled CCD camera for recording the spectra

3. Results and discussions

Fig. 1 shows TSC spectra recorded at a heating rate of 0.15 K/s for samples with different Gd concentrations (N_{Gd}). A broad feature peaking at around 50 K (TSC-1) is visible in all samples, while an additional broad feature centered around 120 K (TSC-2) is present only in Gd doped samples. Fig. 1(e) shows the TSC spectra recorded at different heating rates for sample D. These measurements are carried out under 0.2 V of applied bias. Here, for the shake of clarity, only the edge of TSC-2 feature is shown in an expanded scale. Evidently, the peak shifts to higher temperatures as the heating rate (β) increases. In fact, TSC-1 have also been found to shift to higher temperatures with the increase of β. This

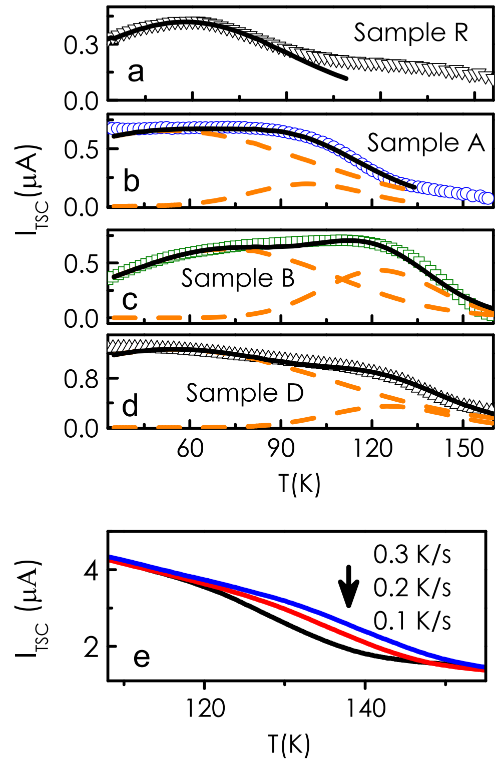

Fig. 1. (Color online) TSC spectra recorded at a heating rate β=0.15 K/s for the (a) undoped sample R and Gd doped samples (b) A, (c) B and (d) D. In each panel, solid black line represents the fitting of the TSC profile and orange dashed lines stand for the simulated TSC spectra for individual trap distributions. (e) TSC spectra recorded for different heating rates for sample D.

confirms that the origin of these peaks is certain types of trap states.

Thermally stimulated current (I_{TSC}) can be expressed as [20]

$$I_{TSC} = e\mu_n\tau_n V\frac{wd}{l}N_T e_n \exp\left(-\int_{T_o}^{T}\frac{e_n}{\beta}dT'\right) \quad (1)$$

where e is the electronic charge, μ_n and τ_n the carrier mobility and lifetime, V the bias voltage,w the width of the contact pads, l the separation between the pads and d the thickness of the layer. N_T the trap concentration and T_o the trap filling temperature. $e_n=\sigma_n v_{th}(g_o/g_1)N_C T^{3/2}\exp(-E/k_BT)$ the thermal escape rate for the carriers from the trap, where g_o/g_1 the degeneracy factor and $v_{th}=\sqrt{8k_BT/\pi m^*}$ the average thermal velocity of the carriers, m^* the carrier effective mass, E the position of the trap with respect to the band edge (conduction/valence band edge for electron-/hole-traps) and $N_C T^{3/2}$ the effective density of states. σ_n the capture cross section associated with the trap. Now, $e_n=\sigma_n\sqrt{8k_B/\pi m^*}(g_o/g_1)N_C T^2\exp(-E/k_BT)$. It should be noted that the density of states (DOS) distribution for a given trap is usually assumed to be a delta function. However, keeping in mind the wider nature of both the TSC features observed in these samples as compared to those are reported for GaN elsewhere [22,20], a Gaussian distribution for the thermal activation energy E has been considered for each type of traps. This means that N_T can be expressed as $N_T(E)=N_T^o\exp[-(E-E_o)^2/2c^2]$, where E_o is the center of the distribution and c the standard deviation of the distribution. Eq. (1) can now be expressed as

$$I_{TSC} = \Omega T^2\int_0^\infty \exp\left[-\frac{(E-E_o)^2}{2c^2}-\frac{E}{k_BT}\right] \times\exp\left[-\frac{\Gamma}{\beta}\int_{T_o}^{T}T'^2\exp\left(\frac{E}{k_BT'}\right)dT'\right]dE \quad (2)$$