Quantum coherence of electrons in random networks of c-axis oriented wedge-shaped GaN nanowalls grown by molecular beam epitaxy

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Abstract
The depth distribution of the transport properties as well as the temperature dependence of the low field magneto-conductance for c-axis oriented GaN nanowall network samples grown with different average wall-widths ($t_{av}$) are investigated. Magneto-conductance recorded at low temperatures shows clear signature of weak localization effect in all samples studied here. The scattering mean free path $l_e$ and the phase coherence time $\tau_{ph}$ are extracted from the magneto-conductance profile. Electron mobility estimated from $l_e$ is found to be comparable with those estimated previously from room temperature conductivity data for these samples, confirming independently the substantial mobility enhancement in these nanowalls as compared to bulk. Our study furthermore reveals that the high electron mobility region extends down to several hundreds of nanometer below the tip of the walls. Like mobility, phase coherence length ($l_{ph}$) is found to increase with the reduction of the average wall width. Interestingly, for samples with lower values of the average wall width, $l_{ph}$ is estimated to be as high as 60 $\mu$m, which is much larger than those reported for GaN/AlGaN heterostructure based two-dimensional electron gas (2DEG) systems.

Keywords: GaN, nanowall network, quantum coherence, weak localization

(Some figures may appear in colour only in the online journal)

1. Introduction
Recently, it has been found that networks of GaN nanowalls show certain interesting structural, optical and transport properties [1–7]. Electron mobility in c-axis oriented wedge-shaped GaN nanowall networks grown on sapphire substrates is estimated to be several orders of magnitude larger [6] than that is observed in GaN bulk [8]. This observation is consistent with a very high lateral conductivity reported in Si doped GaN nanowall networks grown on Si (111) substrate by Zhong et al [3] Interestingly, mobility has been found to increase with the reduction of the average width of the walls [7]. The origin of these effects are not very clear yet. High mobility was speculated to be resulting from the transport of electrons through the edge states located at the top edges of the walls [6]. It is, thus, important to find out, whether the effect is indeed confined only at the tip of the walls or not. Furthermore, keeping in mind that the system exhibits very high electron mobility (estimated to be as high as $\approx 10^4$ cm$^2$ Vs$^{-1}$), it will be interesting to study the quantum interference effect here. One of the important parameters of quantum interference is the phase coherence length ($l_{ph}$), that is the average length scale over which the phase information of an electron remains intact. In fact, there are many device proposals utilizing quantum interference [9].
However, device size has to be less than $l_\phi$, which is one of the key factors for the realization of these devices. Weak localization effect (WL) is often utilized to estimate $l_\phi$ [10, 11]. WL is a negative correction to the conductance arises due to the enhancement of the effective scattering cross-section as a result of constructive quantum interference between a closed path of electron and its time reversed path. When a magnetic field is applied, it reduces the negative correction by removing the phase coherence between the two paths [10], resulting in a positive magneto-conductance at low fields.

Here, we have investigated quantum coherence effect in several c-axis oriented GaN nanowall network samples grown with different average wall-widths ($t_{av}$) by studying the temperature dependence of the low field magneto-conductance. Weak localization effect is observed in all nanowall samples studied here. Two important parameters, namely the scattering mean free path $l_s$ and the phase coherence time $\tau_\phi$, are extracted by fitting the magneto-conductance data using a theory proposed by Beenakker and Houten [12]. Electron mobility estimated from $l_s$ is found to be comparable with those estimated previously from room temperature conductivity data for these samples [6, 7] confirming independently the substantial mobility enhancement in these nanowalls as compared to bulk. Like mobility, phase coherence time also shows an enhancement with the reduction of the average wall width. Interestingly, for an average wall width of 10 nm, the mobility is still significantly fine only at the tip of the walls. Rater, it extends down to several hundreds of nanometer below the tip.

2. Experimental section

Nanowall network samples investigated here were grown directly on c-plane sapphire substrates using plasma assisted molecular beam epitaxial (PA-MBE) with different growth conditions. The Ga and N$_2$ fluxes were kept constant at 3.86 $\times$ 10$^{14}$ cm$^{-2}$ s$^{-1}$ and 4.5 sccm, respectively. Only the substrate temperature and the duration of growth were varied. Further details about the growth, structural, electrical and luminescence properties of these samples can be found elsewhere [1, 6, 7]. The average width of the walls ($t_{av}$) for all of our nanowall network samples was estimated by fitting, with a Gaussian, the line scan profiles taken at random locations of top view SEM images recorded over different parts of the surface. [7] The results are given in table 1. The average electron concentration ($n$) was obtained by measuring the thermoelectric power at 300 K for these samples. [6] Finally, the electron mobility [$\mu_e$(300 K)] was obtained from the conductivity and electron concentration data following the model proposed in [6]. The values for $n$ and $\mu_e$(300 K) thus obtained are also listed in table 1. Magnoeto transport measurements were carried out in a liquid He-cryostat equipped with a superconducting magnet in the temperature range of 1.8 K to 5 K. Magnetic field was varied between $\pm$500 Oe. Indium contact pads, which result in good ohmic contacts, were fabricated on the sample surface. Measurements were performed in four probe van-der-pauw contact geometry. 4M aqueous solution of KOH is used to etch the nanowalls for different time durations to study the depth distribution of their transport properties.

3. Results and discussion

Figure 1 shows the top view scanning electron microscope (SEM) images for sample F before (figure 1(a)) and after 50 min of etching (figure 1(b)). Evidently, the walls are forming a well connected network structure, which remains intact even after the KOH treatment. It is clearly from the insets that the height of the walls is reduced from 1.2 $\mu$m to 600 nm after the treatment. The histograms representing the distribution of the tip width of the walls, which are obtained by analyzing line scan profiles on several SEM top view images recorded at different parts of the sample, are shown for the sample before (figure 1(c)) and after 50 min of etching (figure 1(d)). The peak of the profile clearly shifts from $\pm$15 nm prior to the dipping to $\pm$40 nm after dipping. This finding also suggests a wedge-shaped structure of the nanowalls. The insets are showing the cross-sectional SEM images for the respective samples.

Table 1. The average wall width $t_{av}$, carrier concentration $n$ and electron mobility $\mu_e$ are listed for these samples.

| Sample | $t_{av}$ (nm) | $n$ ($10^9$ cm$^{-2}$) | $\mu_e$ (cm$^2$ V$^{-1}$ s$^{-1}$) |
|--------|---------------|-------------------------|-------------------------------|
| A      | 10 ± 5.9      | 1.25                    | 2.05 $\times$ 10$^4$          |
| B      | 12 ± 3.3      | 1.19                    | 1.01 $\times$ 10$^4$          |
| C      | 15 ± 3.8      | 1.23                    | 1.1 $\times$ 10$^4$          |
| F-1$^a$| 18 ± 5.3      | 1.17                    | 7.9 $\times$ 10$^2$          |
| D      | 40 ± 10       | 1.30                    | 5.5 $\times$ 10$^2$          |
| E      | 60 ± 10       | 1.32                    | 28                            |

$^a$Sample F after 50 min of etching.
field (negative magneto-resistance) is clearly visible at low fields. This can be attributed to the weak localization effect (WL). It is noticeable that around $B = B_0$, where $t_{av}$ is only 10 nm, $\Delta G(B)/G(0)$ changes much faster than that of sample C and D with $t_{av} \approx 20$ and 60 nm, respectively. In fact, $\Delta G(B)/G(0)$ increases more rapidly as the average wall width decreases. Right inset of the figure schematically represents the direction of the external magnetic field $B$ and current $I$ with respect to the orientation of the walls.

Figure 4 compares the $\Delta G(B)/G(0)$ profiles recorded at 2 K for sample F before and after etching. In both the cases, weak
localization effect is quite evident. $\Delta G(B)/G(0)$ changes more rapidly for the unetched sample than that for the etched one. In fact, the rapidity at which $\Delta G(B)/G(0)$ changes is found to decrease as the duration of etching increases. This observation is in accordance with that of figure 3, noting that $t_e$ has been found to increase with etching time.

Figure 5 shows $\Delta G(B)/G(0)$ profiles recorded at different temperatures for sample A. It is noticeable that $\Delta G(B)/G(0)$ increases sharply up to a magnetic field $B_c$ (marked by arrows in the figure), beyond which the rate abruptly falls. Similar results are obtained for other samples as well (position of $B_c$ is marked by arrows in figures 3 and 4 for other samples). Interestingly, $\Delta G(B)/G(0)$ decreases with the increase of temperature and finally above 3 K, it becomes so small that hardly can now be the effective number of parallel channels connecting the contact pads (see the right inset of figure 3). $N_{ch}$, which should now be the number of effective parallel channels connecting the contact pads, can thus be larger than one. It should be noted that the weak antilocalization has been reported in Group III- nitride heterostructure based 2DEG systems [13–15], where it is attributed to spin-orbit coupling arising through Rashba mechanism [13]. These results thus suggest that the spin-orbit coupling is insignificant in these samples. Note that neither weak localization nor weak antilocalization has so far been observed in bulk phase of these materials. Since the magnetic field is applied perpendicular to the sample surface in magnetoresistance measurements, all the walls are subjected to a magnetic field, which is acting parallel to their heights as schematically shown in the inset of figure 3. Conductance $G$ at a given magnetic field $B$ can be expressed as $G(B) = G_0 + \Delta G^{wl}(B)$, where $\Delta G^{wl}(B)$, the weak localization correction. The change in conductance $\Delta G(B) = G(B) - G(0) = \Delta G^{wl}(B) - \Delta G^{wl}(0)$.Beenakker and Houten have shown that for a thin layer, which is subjected to a magnetic field acting parallel to the layer plane, the quantum correction to the conductance as a function of the magnetic field can be expressed as [12]

$$
\Delta G^{wl}(B) = -N_{ch} e^2 B / (2\pi^2 \hbar) \ln \left[ \left( \frac{l_e + l_{ch}}{\tau_{\phi}} \right)^{-1} + 1 \right] \tag{1}
$$

where $\tau_p$ is the mean free time between two elastic collisions, $\tau_p$ the phase coherence time and $\tau_{\phi}$ the magnetic field dependent phase coherence time. $l_e = C_l B / v_f (w/v_f)$ in weak magnetic field regime $l_e \gg \sqrt{w/v_f}$, where $C_l = 16$ and 12.1 for diffuse and specular surface scattering, respectively, $v_f$ the fermi velocity, $l_e = \xi_f v_f$ the mean free path, $l_{ch} = (\hbar/eB)^{1/2}$ the magnetic length and $w$ the thickness of the layer. The prefactor $N_{ch} = 1$ for a single film. However, in case of a network of walls, one should consider multiple parallel channels connecting the contact pads. The estimated values of mobility from conductance measurements as given in table 1 suggest that for all nanowall network samples except sample D and the sample F after etching (Sample F-1), $l_e \gg w$ condition is satisfied. Sample D and F-1 belong to the dirty metal regime $l_e \ll w$, where $t_e$ is given as $t_e = 4h^2/w^2e^2B^2D$. [12] Here, $D$ is the diffusion coefficient, which is given for a thin film with diffuse boundary scattering as $D = \frac{1}{3} l_e \frac{1}{2w} \int_{-w}^{w} \int_{0}^{\infty} \sqrt{x(1-x^2)}(1 - \exp(-w/2l_e)dx)$ [16, 17].

Difference in conductance $\Delta G(B) = G(B) - G(0)$ for $l_e \gg w$ can now be expressed as

$$
\Delta G^{wl}(B) = -N_{ch} e^2 B / (2\pi^2 \hbar) \ln \left[ \left( \frac{l_e}{\tau_{\phi}} + \frac{l_{ch}^3 B^2}{C_l^2 \hbar^2} \right)^{-1} + 1 \right] \tag{2}
$$

where $l_e = \frac{1}{2w} \int_{-w}^{w} \int_{0}^{\infty} \sqrt{x(1-x^2)}(1 - \exp(-w/2l_e)dx)$.
Table 2. The effective number of channels connecting between two Indium metal pads \(N_b\), elastic mean free path \(l_e\), elastic mean time \(t_e\), fermi velocity \(v_f\), electron mobility estimated from \(l_e\) and \(v_f\) using \(\mu\text{MR}(2\, \text{K}) = e l_e / m^* v_f\), phase coherence time \(\tau_\phi\) and phase coherence length \(l_\phi\) are given.

| Sample | \(N_b\) (cm\(^{-3}\)) | \(l_e\) (nm) | \(t_e\) \(10^{-12}\) (s) | \(v_f\) \(10^5\) (M s\(^{-1}\)) | \(\mu\text{MR}(2\, \text{K})\) (cm\(^2\) V s\(^{-1}\)) | \(\tau_\phi\) \(10^{-10}\) (s) | \(l_\phi\) (\(\mu m\)) |
|--------|-----------------|-------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| A \(^{a}\) | 1400 | 2080 \(\pm\) 208 | 4.96 | 4.19 | 43 590 | 6080 \(\pm\) 608 | 60.6 \(\pm\) 6.1 |
| B | 2750 | 1130 \(\pm\) 113 | 2.8 | 4.05 | 24 524 | 945 \(\pm\) 95 | 23.8 \(\pm\) 2.4 |
| C | 3000 | 723 \(\pm\) 72.3 | 1.78 | 4.05 | 15 646 | 714 \(\pm\) 71 | 21.6 \(\pm\) 2.2 |
| D | 4000 | 28 \(\pm\) 2.8 | 0.07 | 4.045 | 610 | 1.96 \(\pm\) 0.6 | 0.65 \(\pm\) 0.07 |
| F-1 \(^{b}\) | 3000 | 25 \(\pm\) 2.5 | 0.06 | 4.08 | 549 | 1.25 \(\pm\) 0.13 | 0.57 \(\pm\) 0.06 |
| D | 4410 | 1 \(\pm\) 0.1 | 0.003 | 4.053 | 24 | 6.2 \(\pm\) 0.62 | 0.30 \(\pm\) 0.03 |

\(^{a}\) sample F after 50 min of KOH etching, and
\(^{b}\) values for sample A calculated at 2.4 K.

and for \(l_e \ll w\) (dirty metal regime) as

\[
\Delta G^{\text{el}}(B) = -N_b e^2 \frac{e^2}{2\pi^2 h} \left[ \ln \left( \frac{l_e}{v_f \tau_\phi} + \frac{w^2 \alpha_{DB}^2}{4v_f h^2} \right) + 1 \right] - \ln \left( \frac{l_e}{v_f \tau_\phi} + 1 \right)
\]

Figure 6. Compares the variation of phase coherence time \(\tau_\phi\) as a function of temperature for different samples.

Experimental data of \(\Delta G(B)\) are fitted using equation (2) for samples B, C, F and equation (3) for sample D and F-1 with \(N_b\), \(\tau_\phi\) and \(l_e\) as fitting parameters. Here, the boundary scattering is considered to be diffusive, meaning the value of \(C_1\) is taken to be 16, \(w\) is considered to be equal to \(l_\text{av}\). Note that \(v_f = \frac{\hbar}{m^* \sqrt{2m_n e}}\) for a two dimensional carrier gas, where the areal concentration of carriers is \(n_t\). Hence, \(n_t = nt_\text{av}\) and hence \(v_f\) is estimated from 3D electron concentration \(n\) determined from thermoelectric power measurements (see table 1) for these samples. Results of the fitting for different samples are shown in figures 3 and 4. The best fit values obtained at 2 K for all the samples are listed in table 2. Electron mobility \(\mu\text{MR}(2\, \text{K})\), which has been estimated from \(l_e\) and \(v_f\) through \(\mu\text{MR}(2\, \text{K}) = e l_e / m^* v_f\) with \(m^* = 0.2m_e\) being the electron effective mass for GaN, is also listed in the table. Note that these mobility values are quite comparable with the values estimated previously from room temperature conductivity data for these samples (see table 1) [6, 7]. Moreover, \(\mu\text{MR}(2\, \text{K})\) increases as \(t_\text{av}\) decreases, which is again in agreement with our previous observation [6, 7]. These findings not only verify the consistency of this fitting but also confirm independently the substantial increase of mobility in nanowalls as compared to bulk specially for samples with lower wall-widths. Another noticeable point is the observation of significantly large phase coherence time \(\tau_\phi\) particularly for thinner walls. For instance, in sample A, \(\tau_\phi\approx 600\, \text{ps}\), which is much higher than those reported for GaN/AlGaN heterostructure 2DEG, suggesting that the inelastic scattering rate is significantly lower in this sample as compared to heterostructure 2DEG systems [13–15]. The phase coherence length \(l_\phi\) is also estimated following the relation \(l_\phi = \sqrt{\tau_\phi D}\). Evidently, \(l_\phi\) also increases as \(t_\text{av}\) decreases. Note that for small values of \(t_\text{av}\), \(l_\phi\) becomes as long as 60 \(\mu m\), which is higher than that is estimated in a narrow channel of GaN/AlGaN heterostructure [15]. It should be mentioned that larger the value of \(l_\phi\) faster will be the variation of \(\Delta G(B)\) with \(B\) around \(B = 0\). \(\Delta G(B)/G(0)\) profiles shown in figures 3 and 4 thus clearly suggests an increase of \(l_\phi\) with the reduction of \(t_\text{av}\).

Figure 6 compares the variation of phase coherence time \(\tau_\phi\) as a function of temperature for different samples. It is evident that \(\tau_\phi\) does not vary much with temperature within 2–3 K range for all samples. This is also clear from almost temperature independent nature of the shape of \(\Delta G(B)/G(0)\) profiles around \(B = 0\) for sample A within that temperature range as shown in figure 5. The observation of figure 5 furthermore suggests that the main reason for the vanishing of peak localization signature with the increase of temperature is the rapid reduction of the amplitude of the quantum correction, not the decrease of \(\tau_\phi\). Note that in semiconductors, the major source of dephasing is believed to be the electron-electron scattering, which should result in a monotonic decrease of \(\tau_\phi\) with increasing temperature. It should be mentioned that temperature independence of \(\tau_\phi\) has been reported in other systems such as narrow channels of GaAs/InGaAs heterostructures [18] and GaAs nanowires [19]. The mechanism has been ascribed to certain surface scattering process [20] and also to spin–spin scattering by residual magnetic impurities.
It is interesting to note that $\tau_p$ increases as $t_{av}$ decreases. This could mean that the rate of inelastic scattering, which governs $\tau_p$, increases with $t_{av}$. Note that mobility is also found to increase with the decrease of $t_{av}$ for these samples. The observation of such a significantly high mobility and inelastic scattering time might suggest a quantum confinement of electrons in the walls. Note that the quantum confinement of carriers in a 2D channel can enhance the mobility by several orders of magnitude due to the decrease in scattering cross-section as a result of reduced dimensionality. Confinement in this case is likely to be 2D in nature as the high electron mobility region is found to extend down to several hundreds of nanometer below the tip of the walls (figure 1). It is plausible that the accumulation of negative charges on the nanowall facets can result in a 2D confinement of electrons in the wall. These charges might be pushing the electrons inward leading to positive depletion regions at the boundaries and a 2D quantum confinement in the central plane parallel to the height of the walls shown schematically in figure 7. Reduction of elastic as well as inelastic scattering rates with $t_{av}$ can be explained in terms of the enhancement of the 2D confinement as $t_{av}$ decreases.

4. Conclusions

In conclusion, we have studied the depth distribution of the transport properties as well as the temperature dependence of the low field magneto-conductance at low temperatures for several c-axis oriented GaN nanowall network samples grown with different average wall-widths ($t_{av}$). Weak localization effect is observed in all nanowall samples studied here. Scattering mean free path $l_e$ and the phase coherence time $\tau_p$ are extracted by fitting the magneto-conductance data using a theory proposed by Beenakker and Houten. Electron mobility estimated from $l_e$ is found to be comparable with that is estimated previously from room temperature conductivity data for these samples [7], not only verifying the consistency of this fitting but also confirming independently the substantial enhancement of mobility in these nanowalls as compared to bulk. Interestingly, for samples with smaller wall widths, $\tau_p$ is estimated to be as high as 60 $\mu$m, which is much larger than those reported for GaN/AlGaN heterostructure based two dimensional electron gas (2DEG) systems. Our study, furthermore, reveals that the high electron mobility region does not confine at the tip of the walls. Rather, it extends down to several hundreds of nanometer below the tip. Both $l_e$ and $\tau_p$ are found to be increased with the decrease of the average wall width ($t_{av}$), suggesting a reduction of elastic as well as inelastic scattering rates with $t_{av}$.

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References

[1] Kesaria M, Shetty S and Shivaprasad S M 2011 Cryst. Growth. Des. 11 4900
[2] Kesaria M and Shivaprasad S M 2011 Appl. Phys. Lett. 99 143105
[3] Zhong A and Hane K 2012 Nanoscale Res. Lett. 7 686
[4] Zhong A and Hane K 2013 Japan. J. Appl. Phys. 52 08JE13
[5] Zhong A, Sasaki T and Hane K 2014 Int. J. Hydrogen. Energy 39 8564
[6] Bhasker H P, Dhar S, Sain A, Kesaria M and Shivaprasad S M 2012 Appl. Phys. Lett. 101 132109
[7] Bhasker H P, Thakur V, Kesaria M, Shivaprasad S M and Dhar S 2014 AIP Conf. Proc. 1583 252
[8] Dhar S and Ghosh S 1999 J. Appl. Phys. 86 2668
[9] Datta S, Melloch M R, Bandyopadhyay S and Lundstrom M S 1986 Appl. Phys. Lett. 48 487
[10] Bergmann G 2010 Int. J. Mod. Phys. B 24 2015
[11] Kramer B and MacKinnon A 1993 Rep. Prog. Phys. 56 1469
[12] Beenakker C W J and vanHouten H 1988 Phys. Rev. B 38 3232
[13] Schmult S, Manfra M J, Punnoose A, Sergent A M, Baldwin K W and Molnar R J 2006 Phys. Rev. B 74 033302
[14] Thillosen N, Schüpers T, Kaluza N, Hardtdegen H and Guzenko V A 2006 Appl. Phys. Lett. 88 022111
[15] Lehnen P, Schüpers T, Kaluza N, Thillosen N and Hardtdegen H 2007 Phys. Rev. B 76 205307
[16] Fuchs K 1938 Proc. Camb. Phil. Soc. 34 100
[17] Sondheimer E H 1952 Adv. Phys. 1 1
[18] Hiramoto T, Hirakawa K, Iye Y and Ikoma T 1989 Appl. Phys. Lett. 54 2103
[19] Hansen A E, Björk M T, Fasth C, Thelander C and Samuelson L 2005 Phys. Rev. B 71 205328
[20] Taylor R P, Whittington M L, Matin P C, Beaumont S P, McIntyre I, Thomas S and Wilkinson C D W 1988 Surf. Sci. 196 52
[21] Lin J J and Giordano N 1987 Phys. Rev. B 35 1071
[22] Bhasker H P, Thakur V, Shivaprasad S M and Dhar S (arXiv:1410.1295)