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Anomalous temperature dependence of the electrical resistivity in $In_{0.17}Ga_{0.83}N$

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ABSTRACT

Resistivity and Hall effect measurements on n-type undoped $In_{0.17}Ga_{0.83}N$ alloy grown by metal-organic vapor phase epitaxy (MOVPE) technique were carried out as a function of temperature (15–350 K). $In_{0.17}Ga_{0.83}N$ alloy is regarded as a highly degenerate semiconductor system with a high carrier concentration of $\sim 9.2 \times 10^{19}$ cm^{-3}. An anomalous resistivity behavior is observed over the whole temperature range. The temperature dependent resistivity of $In_{0.17}Ga_{0.83}N$ exhibits a metal-semiconductor transition (MST) around 180 K. The temperature coefficient of resistivity is negative at low temperatures ($T < 180$ K) and it becomes positive at relatively high temperatures ($T > 180$ K). In addition to this, a negative magnetoresistivity (MR) has been observed below 180 K. The temperature dependent resistivity of $In_{0.17}Ga_{0.83}N$ alloy is explained in the terms of the electron–electron interaction (EEI) and the weak localization (WL) phenomenon at low temperatures ($T < 180$ K). At high temperatures ($T > 180$ K) the temperature dependent resistivity obeys T^2 law.

1. Introduction

The $In_xGa_{1-x}N$ material system with high Ga composition has attracted a great deal of attention in recent years. It is used as an active layer in blue and green light emitting diodes and lasers [1–4]. In spite of numerous studies having been carried out on the optical and structural properties of $In_xGa_{1-x}N$ [5,6], studies on the electrical transport properties of this material system are still limited due to difficulty in controlling the conductivity. Since the growth temperature of $In_xGa_{1-x}N$ is generally lower than that of GaN, the decomposition rate of ammonia (NH_3) becomes low at low-growth temperatures [7]. The low temperature growth conditions cause an increment in number of nitrogen vacancies. Nitrogen vacancies increment results a high background carrier concentration in undoped $In_xGa_{1-x}N$ alloy. At sufficiently high impurity concentrations, the electron transport in semiconductors exhibits metallic behavior above the critical Mott concentration (n_c). In fact, impurity conduction may become significant even at high temperatures in high energy gap Ga-rich $In_xGa_{1-x}N$ alloys in which compensation is nearly full.

As can be seen from the literature, InN and related alloys can exhibit metallic impurity band conduction with a high carrier concentration and in this case they have low mobility values. For the InN with electron concentrations of more than 1×10^{19} cm^{-3}, mobility is normally less than 250 cm^2/Vs [8,9]. Lin et al. [10] observed that the electron concentration in $In_xGa_{1-x}N$ films is temperature independent over a wide temperature range 4 K $\leq T \leq$ 285 K. Their experimental results demonstrated that the electron transport in In-rich $In_xGa_{1-x}N$ films with a carrier concentration $\sim 2 \times 10^{19}$ cm^{-3} show metallic behavior. Geerstet al. [11] studied carrier transport phenomena in n-type $In_xGa_{1-x}N$ with variable temperature (1.7–400 K) and magnetic field (0–30 T) Hall measurements. Their $In_{0.18}Ga_{0.82}N$ sample ($n \sim 4 \times 10^{19}$ cm^{-3} and $\mu \sim 3$ cm^2/Vs) showed metallic conduction, which due to intrinsic shallow donors in $In_xGa_{1-x}N$ material. They also observed that the $In_{0.18}Ga_{0.82}N$ shows a negative magnetoresistance over a wide temperature range (2–350 K) [11].

In the metallic side of the metal-insulator transition, the resistivity of a highly degenerate semiconductor decreases with decreasing temperature as is typical for a good metal. However, the resistivity of a highly degenerate semiconductor can exhibit anomalous behavior at low temperatures, as structural or compositional disorder increases in the material. In such a material, the mean free path between collisions becomes small and quantum effects become important. If the resistivity of highly degenerate semiconductors increases with decreasing temperature at low temperatures, the Boltzmann approach may not successfully describe the transport properties of the material [12]. This requires quantum corrections to the Boltzmann conductivity. It is widely accepted that the electron transport properties of such materials can

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be explained with weak localization (WL) and electron–electron interaction (EEI) phenomenon at low temperatures [12].

In a previous work [6], we showed that $In_xGa_{1-x}N$ layers have a high bowing parameter ∼3.6 eV in the composition range between $x = 0.060$ and $x = 0.105$, which indicates presence of disorder in the structure of $In_xGa_{1-x}N$. Also, in another paper [13], it has been shown that impurity band conduction dominates in electron transport of high degenerate $In_xGa_{1-x}N$ samples ($0.06 \leq x \leq 0.135$) in a temperature range 15–350 K and the temperature dependent the conductivity can be well explained by the model that takes into account EEI and WL [13].

In order to better understand the effect of the increase in carrier concentration on electrical properties of InGaN alloys, another samples exhibiting a metal-semiconductor transition (MST) have been investigated in this work apart from a previous one [13]. We report the measurements of the electrical resistivity in n-type undoped $In_{0.17}Ga_{0.83}N$ alloy grown by metal-organic vapour phase epitaxy (MOVPE) in a temperature range of 15–350 K. Temperature dependent resistivity shows a MST around 180 K. The observed anomalous temperature behavior of resistivity is analyzed using the conventional metallic conduction model ($\rho(T) = \rho_0 + AT^n$) at high temperatures ($T > 180$ K), and in terms of the WL and EEI at low temperatures ($T < 180$ K).

2. Experimental

The $In_{0.17}Ga_{0.83}N$ epilayer presented in this work were grown in an atmospheric pressure cold-wall vertical MOVPE reactor with a shower head configuration. The standard heterostructure included a 80–100 nm thick GaN buffer grown at 510 °C, a 600 nm thick GaN layer deposited at 1080 °C (typical V/III ratio was about 7000), and an $In_xGa_{1-x}N$ alloy deposited at 800 °C under different conditions, in order to vary In content. Details of the growth procedures were given elsewhere [14]. Summary of growth conditions of InGaN alloys are also presented in Table S1 (Supporting information). The In composition was studied by X-ray diffraction (standard $\theta/2\theta$ diffractometer) assuming that the lattice parameter varies linearly with the In fraction according to Vegard's law.

For resistivity and Hall effect measurements by the van der Pauw method, square shaped (5×5 mm^2) sample was prepared with four contacts in the corners. Using annealed indium dots, ohmic contacts to the sample were prepared and their ohmic behavior was confirmed by the current–voltage characteristics. Measurements were made at temperature steps over temperature range 15–350 K using a Lake Shore Hall effect measurement system (HMS). At each temperature step, the Hall coefficient (with maximum 5% error) and resistivity (with maximum 0.2% error in the studied range) were measured for both current directions, both magnetic field directions that were perpendicular to the surface and all the possible contact configurations at 14 magnetic field steps between 0 and 1.4 T (with 0.1% uniformity).

3. Results and discussion

Fig. 1 and 2 show the resistivity and carrier concentration and mobility variation of the $In_{0.17}Ga_{0.83}N$ alloy as a function of temperature between 15 and 350 K, respectively. The unintentionally doped $In_{0.17}Ga_{0.83}N$ alloy exhibits n-type conductivity, which may be due to the presence of nitrogen vacancies. The temperature dependent of the resistivity of $In_{0.17}Ga_{0.83}N$ exhibits a semiconducting behavior (resistivity decreases with temperature) at low temperatures ($T < 180$ K). Then it reaches a minimum where metallic behavior starts to appear, i.e. the resistivity increases as the temperature increases at high temperatures ($T > 180$ K). Result of the resistivity measurements indicates that a MST occurs around 180 K in $In_{0.17}Ga_{0.83}N$ alloy.

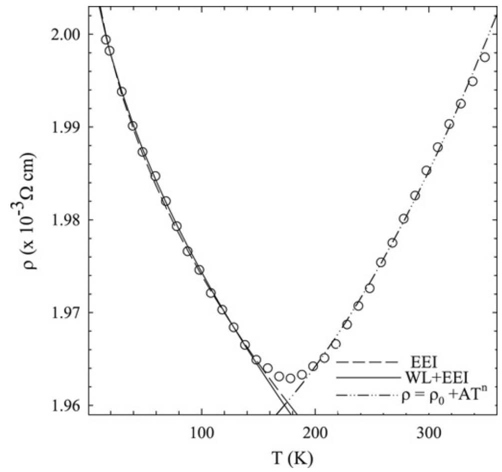

Fig. 1. Temperature dependence of the resistivity (ρ) for $In_{0.17}Ga_{0.83}N$ in a temperature range of 15–350 K. Open circles show the experimental points, solid line is the best fit with Eq. (1) which includes both the WL and the EEI and dashed line is the best fit with Eq. (1) without the WL term for $T < 180$ K, while the dotted line is the best fit with Eq. (6) for $T > 180$ K.

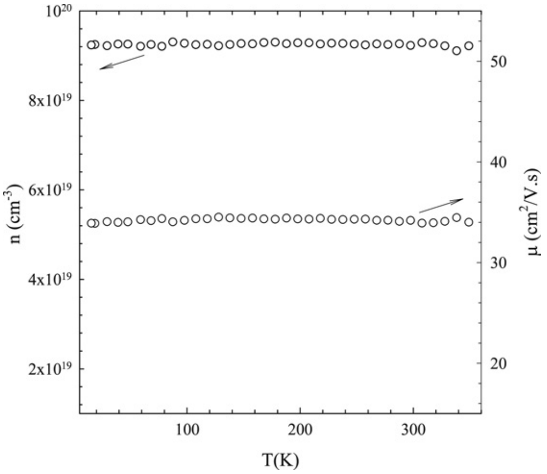

Fig. 2. Temperature-dependent Hall mobility (μ) and carrier density (n) for $In_{0.17}Ga_{0.83}N$ in a temperature range of 15–350 K.

The $In_{0.17}Ga_{0.83}N$ has a carrier concentration of 9.2×10^{19} cm^{-3} with mobility of 34 cm^2/Vs at room temperature. This electron concentration value is much higher than the critical carrier concentration of 7.7×10^{17} cm^{-3} for the Mott transition in $In_{0.17}Ga_{0.83}N$ respect to the previously reported $In_xGa_{1-x}N$ samples ($0.06 \leq x \leq 0.135$) [13]. When the carrier concentration exceeds the critical carrier concentration, the increased Coulomb interaction leads to a reduction in mobility and, consequently, an increase in resistivity as temperature increases, which is a characteristic of metallic behavior [15]. The Hall carrier concentration of $In_{0.17}Ga_{0.83}N$ is nearly temperature independent over the whole studied temperature range. A metallic state in $In_{0.17}Ga_{0.83}N$ has been obtained, as evidenced by an estimated room-temperature conductivity of 504 $(\Omega$ cm$)^{-1}$, nearly temperature-independent resistivity and a very high carrier concentration. Observation of low mobility values can be explained with residue imperfection scattering in the $In_xGa_{1-x}N$ [10], since values of the resistivity and mobility at 15 K are 10% smaller than their values at 350 K in $In_{0.17}Ga_{0.83}N$ alloy. Nitrogen vacancies and the large concentration of dislocations due to the large lattice mismatch between GaN and $In_xGa_{1-x}N$, can affect the electrical properties of the sample and it may result a decrease in the mobility. Wanget al. [16] have carried out a detailed mobility analysis for $In_xGa_{1-x}N$ alloys. In order to study the scattering mechanism in $In_xGa_{1-x}N$ alloys, a simple mobility fit was made by assuming that the main scattering mechanisms are due to ionized impurity ($\mu_{II}(T)\alpha T^{3/2}$), alloy ($\mu_{al}(T)\alpha T^{-1/2}$) and optical phonon ($\mu_{op}(T)\alpha T^{-3/2}$) mechanisms, and it was observed that the agreement between experimental data of mobility and fit is excellent [16]. When we tried to fit our data using these mechanisms as well as space-charge scattering ($\mu_{sc}(T)\alpha T^{-1/2}$) mechanism, we did not obtain any acceptable fit for our highly degenerate $In_{0.17}Ga_{0.83}N$ sample.

We have also failed to fit the resistivity data of $In_{0.17}Ga_{0.83}N$ to the Arrhenius plot ($\rho(T) = \rho(0)\exp(E_a/k_BT)$, where E_a is the activation energy) for $T < 180$ K. This suggests that a simple thermal activation process in temperature range of $T < 180$ K does not dominate the electrical conduction in $In_{0.17}Ga_{0.83}N$. According to Figs. 1 and 2, it can be clearly seen that electron transport is controlled by the impurity band conduction in $In_{0.17}Ga_{0.83}N$. Therefore, we must use another model to explain this semiconducting behavior in $In_{0.17}Ga_{0.83}N$. The MST appears with a carrier concentration of 9.2×10^{19} cm^{-3} in $In_xGa_{1-x}N$, and it completely disappears with carrier concentrations that are close to the critical carrier concentration [13]. In a previous work [13], it showed that the temperature dependent conductivity of degenerate $In_xGa_{1-x}N$ samples ($0.06 \le x \le 0.135$) displaying semiconducting behavior $d\rho/dT < 0$ in the whole temperature range (15–350 K) and it can be well explained by the model that takes into account electron–electron interactions and weak localization [13]. However, it is clear that this model is valid for $In_{0.17}Ga_{0.83}N$ only at low temperatures ($T < 180$ K). So, we cannot apply to this model the resistivity data of $In_{0.17}Ga_{0.83}N$ over the whole measuring temperature range as in $In_xGa_{1-x}N$ samples ($0.06 \le x \le 0.135$). In case of such metallic impurity band conduction, the electron transport may not be explained by the Boltzmann approach [12]. This situation clearly confirms the quantum corrections requirement to resistivity at low temperatures ($T < 180$ K). The quantum correction to resistivity due to WL and EEI effects for three-dimensional case in the absence of a magnetic field is given by [12]

$$\rho(T) = \frac{1}{\sigma(0) + mT^{1/2} + BT^{p/2}}, \tag{1}$$

where the $mT^{1/2}$ term arises from electron–electron interactions and the $BT^{p/2}$ term is the correction to the zero-temperature conductivity ($\sigma(0)$) due to localization effects. If electron–electron interactions are dominant, $p = 2$, whereas $p = 3$ if electron-phonon scattering dominates [12].

As is well known, electron–electron interaction and weak localization effect are usually observed at low temperatures, less than 10 K. However, it was reported that these effects could be considered in high band gap semiconductors having very high carrier concentration [12,13]. In our case, $In_{0.17}Ga_{0.83}N$ alloy has a high band gap ($E_g > 2.5$ eV) and it may be a possible observation of these effects at even high temperatures. It was also reported that the metallic impurity band conduction is dominated in the carrier transport up to room temperature for InGaN [7]. Therefore, we are motivated to fit Eq. (1) to the experimental resistivity data of $In_{0.17}Ga_{0.83}N$ for $T < 180$ K. We see that a good agreement can only be obtained when p is close to 2 for $In_{0.17}Ga_{0.83}N$. So, we set the value of p as 2 in Eq. (1). Fig. 1 shows the resistivity versus temperature accompanying the fitted results (solid line) at low temperatures ($T < 180$ K). The values of the parameters in Eq. (1) are obtained as $\sigma(0) = 496.3 \pm 0.195$ $(\Omega\ \text{cm})^{-1}$, $m = 0.9 \pm 0.0053$ $(\Omega\ \text{cm})^{-1}/\text{K}^{1/2}$ and $B = (1.2 \pm 0.033) \times 10^{-2}$ $(\Omega\ \text{cm K})^{-1}$. If the fitting is restricted in lower temperatures ($T < 80$ K), the corresponding values are $\sigma(0) = 496.5$ $(\Omega\ \text{cm})^{-1}$, $m = 0.87$ $(\Omega\ \text{cm})^{-1}/\text{K}^{1/2}$ and $B = 1.35 \times 10^{-2}$ $(\Omega\ \text{cm K})^{-1}$. It is noted that when the fitting is limited in the temperatures below 80 K, we do not obtain considerable changes in the values of fitting parameters. Therefore, we suggest that this model (Eq. (1)) is applicable to high temperature data (up to 180 K) for $In_{0.17}Ga_{0.83}N$. We can expect that the dominant contribution to the resistivity comes from EEI at zero magnetic fields in the $In_{0.17}Ga_{0.83}N$. Neglecting the WL term in Eq. (1), there is still an acceptable fit as seen from Fig. 1. In this case, the value of m (1.2 $(\Omega\ \text{cm})^{-1}/\text{K}^{1/2}$) is slightly increased. However, the correlation coefficient of the fitting straight line is better with WL term than the without WL term in the fit. So, it can be considered that the contribution to the low temperature resistivity of the $In_{0.17}Ga_{0.83}N$ arises both the WL term, having a small contribution on the resistivity, and EEI term. However, the m and B parameters should be calculated theoretically, since a good fit of the measured data is essential but not sufficient criterion for applicability of Eq. (1). The m and B fitting (experimental) parameters should be in agreement with the theoretical values.

In order to determine the m and B parameters theoretically, firstly, we deal with m coefficient. Coefficient m in the EEI contribution to the low-temperature resistivity is given by [12],

$$m = \alpha\left[\frac{4}{3} - \left(\frac{3}{2}\right)\gamma F_\sigma\right], \tag{2}$$

where α is defined as,

$$\alpha = \frac{e^2}{\hbar}\left(\frac{1.3}{4\pi^2}\right)\left(\frac{k_B}{2\hbar D}\right)^{1/2}, \tag{3}$$

where, D is the diffusion coefficient. In order to determine D, the relation ($\sigma(0) = 2S_0\eta e^2 DN(0)$) can be used, where $N(0)$ is the concentration of states ($N(0) = m^* k_F/2\pi^2\hbar^2$). Omitting anisotropy of effective mass, it is assumed $S_0\eta = 1$ for InGaN. γF_σ is the Coulomb interaction parameter where γ is taken as 1.95 for $In_xGa_{1-x}N$ [13]. The quantity F_σ is related to the Fermi-liquid parameter F by [17]

$$F_\sigma = \left(-\frac{32}{3}\right)\left[1 - \frac{3F}{4} - \left(1 - \frac{F}{2}\right)^{3/2}\right]F^{-1}. \tag{4}$$

In general, F values ranges between 0 and 1 in disordered metallic systems [18]. Using the Thomas–Fermi approximation, F can be determined as 0.22 for $In_{0.17}Ga_{0.83}N$. Since the value of calculated γF_σ (0.43) is smaller than 8/9, the m parameter which is calculated from the Eq. (2) is positive and its value is 1.51 $(\text{cm})^{-1}/\text{K}^{1/2}$ for $In_{0.17}Ga_{0.83}N$. The obtained theoretical value of coefficient m is of the same order of magnitude as found in degenerate semiconductor systems [12,13,18].

Now, we consider B coefficient in the WL term in Eq. (1), if it is assumed $p = 2$ to be, B is determined by [19],

$$B = \frac{e^2}{\hbar\pi^2}\left[\frac{S_0\eta}{2}\left(\frac{c}{D}\right)^{1/2}\right]. \tag{5}$$

The coefficient B is calculated as 2.2×10^{-2} $(\text{cm K})^{-1}$ from Eq. (5). Actually, it is clear that the m and B fitting parameters which are obtained experimentally ($m = 0.90$ $(\Omega\ \text{cm})^{-1}/\text{K}^{1/2}$ and $B = 1.2 \times 10^{-2}(\Omega\ \text{cm K})^{-1}$) from Eq. (1) are in reasonable agreement with the calculated values ($m = 1.51$ $(\Omega\ \text{cm})^{-1}/\text{K}^{1/2}$ and $B = 2.2 \times 10^{-2}$ $(\Omega\ \text{cm K})^{-1}$) from Eqs. (2) and (5), respectively. On the other hand, compared with early reported values [12,19,20] for metallic systems, the obtained value of B is very small. Therefore, it can be considered that the WL effect is very small in $In_{0.17}Ga_{0.83}N$.

In order to achieve deeper understanding the low temperature carrier transport properties of the $In_{0.17}Ga_{0.83}N$, we measured the magnetic field dependence of resistivity of the sample at three selected temperatures. Defining the magnetoresistivity as

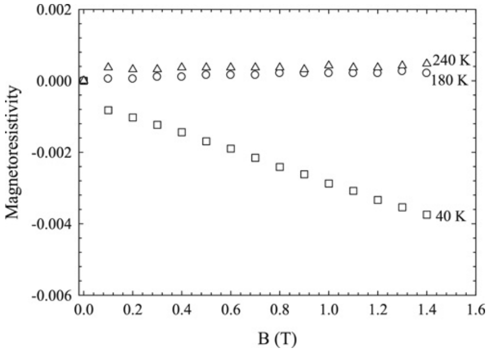

Fig. 3. Magnetoresistivity at three temperatures for $In_{0.17}Ga_{0.83}N$.

MR $= [\rho(B) - \rho(0)]/\rho(0)$, the MR variations with magnetic field at 40 K, 180 K and 240 K are shown in Fig. 3. It is seen that the MR is relatively weak and it becomes negative below 180 K. Arnaudov et al. [21] observed a similar MR behavior in InGaN/GaN quantum well. Since there is quantum interference between the electronic waves at low temperature, the back-scattering probability of electrons will be enhanced. This leads to a result in the terms of weak localization of electrons. WL is suppressed, if an external magnetic field is applied. Because the magnetic field suppresses the wave coherence, thus, quantum interference effects are reduced and the resistivity is decreased. The contribution to the MR due to weak localization is negative [12]. However, the contribution of the WL to electrical conduction of $In_{0.17}Ga_{0.83}N$ is very weak compared with EEI. Ultimately, the semiconducting behavior in $In_{0.17}Ga_{0.83}N$ can be explained well in terms of EEI and WL effects at low temperatures ($T < 180$ K).

Finally, we considered the resistivity data at high temperatures ($T > 180$ K). The temperature dependent resistivity of $In_{0.17}Ga_{0.83}N$ shows a positive temperature coefficient of the resistivity above 180 K, which is a characteristic of metallic behavior. As previously mentioned, due to the extremely high carrier concentration and the increased Coulomb interactions, an increase in resistivity is observed as the temperature increases in $In_{0.17}Ga_{0.83}N$. The metallic conduction observed above MST can be explained by a high carrier concentration and the formation of a degenerate band, which appears in heavily doped semiconductors [22]. However, the intrinsic as well as extrinsic donors should cause to a high carrier concentration in unintentionally doped $In_{0.17}Ga_{0.83}N$ material system. So, the observed metallic conduction in this material system above MST may be explained by a large number of nitrogen vacancies which act as donors and lead to degeneracy. The temperature dependence of the resistivity of in the metallic region is characterized by,

$$\rho(T) = \rho_0 + AT^n \qquad (6)$$

where ρ_0, A, and n are the fitting parameters. The temperature dependence of the resistivity is generally explained by T^n law (n is a positive integer). In the temperatures range of $20 \text{ K} < T < \Theta_D$, ($\Theta_D$ is Debye temperature), $n = 3$–5 when electron-phonon scattering dominates or $n = 2$ when electron–electron scattering dominates [23]. At high temperatures ($T > \Theta_D$), the temperature dependence of resistivity follows a linear dependence on the temperature [23]. The Debye temperature Θ_D is about 1042 K for $In_{0.17}Ga_{0.83}N$ [24]. Eq. (6) is fitted to the resistivity data of $In_{0.17}Ga_{0.83}N$ for $180 \text{ K} < T < 350$ K. The results are given in Fig. 1. The open circles in Fig. 1 are the experimental data and the solid line is the best fitted values. The $r^2 = 0.997$ (r = correlation coefficient) was obtained, which indicates a satisfactory fit. It is found that the best fit is obtained with the parameters of $\rho_0 = 1.948 \times 10^{-3}\ \Omega$ cm, $A = 3.02 \times 10^{-10}\ \Omega \text{ cm K}^{-2}$ and $n = 2.05$. In the case of high carrier concentration ($\sim 10^{20}\text{ cm}^{-3}$), the quadratic temperature dependence of resistivity in $In_{0.17}Ga_{0.83}N$ alloy is mainly result of a significant electron–electron scattering at high temperatures.

4. Conclusion

We have investigated the electrical resistivity on n-type undoped $In_{0.17}Ga_{0.83}N$ alloy grown by MOVPE technique in a temperature range of 15–350 K. Anomalous resistivity behavior has been observed in the investigated sample. The temperature dependent resistivity of $In_{0.17}Ga_{0.83}N$ exhibits a MST while representing a minimum around 180 K. MST is attributed to a high carrier concentration ($\sim 10^{20}\text{ cm}^{-3}$), which may be due to a large number of nitrogen vacancies. At low temperatures ($T < 180$ K), temperature dependent resistivity can be well explained by the EEI and the WL effects, with the parameters that are in an agreement with the theoretical predictions. The sample exhibits negative MR below 180 K, which is evidence of the presence of the WL effect. At high temperatures ($T > 180$ K), the temperature dependent resistivity can be well explained in terms of power law dependence with $n = 2$ (quadratic dependence on temperature). Finally, it can be suggested that a further increment in the carrier concentration of $In_xGa_{1-x}N$ alloys may lead to a completely suppressed semiconducting behavior.

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Appendix. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.ssc.2008.11.026.

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