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Determination of the critical indium composition corresponding to the metal–insulator transition in $In_xGa_{1-x}N$ ($0.06 \leqslant x \leqslant 0.135$) layers

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ABSTRACT

The low-temperature conductivity of $In_xGa_{1-x}N$ alloys ($0.06 \leqslant x \leqslant 0.135$) is analyzed as a function of indium composition (x). Although our $In_xGa_{1-x}N$ alloys were on the metallic side of the metal–insulator transition, neither the Kubo-Greenwood nor Born approach were able to describe the transport properties of the $In_xGa_{1-x}N$ alloys. In addition, all of the $In_xGa_{1-x}N$ alloys took place below the Ioeffe–Regel regime with their low conductivities. The observed behavior is discussed in the framework of the scaling theory. With decreasing indium composition, a decrease in thermal activation energy is observed. For the metal–insulator transition, the critical indium composition is obtained as $x_c = 0.0543$ for $In_xGa_{1-x}N$ alloys.

1. Introduction

Recently, $In_xGa_{1-x}N$ alloys have been attracting a great deal of theoretical and experimental interest because of their applications in light emitting diodes [1] as well as in semiconductor lasers [2]. There have been several attempts to understand the outstanding electronic properties of $In_xGa_{1-x}N$ alloys [3,4]. To the best of our knowledge there are no other works that have determined the critical indium composition for metal–insulator transition (MIT) in $In_xGa_{1-x}N$ alloys. The electrical conductivity of a material can be changed from the insulating phase to the metallic phase depending on the composition, doping, pressure, or strain. This is referred to as MIT [5]. In recent years, the problem of the MIT in disordered systems has become increasingly important. On the metallic side of the MIT, the effect of weak localization and electron–electron interactions is important as structural or compositional disorder increases in $In_xGa_{1-x}N$ alloys [4].

The behavior of electrical conductivity with the carrier concentration at low temperatures above the metallic side of MIT is one of the fundamentals for inferring the roles of disorders and electron–electron interactions in a disordered material. The transition from the insulating phase to the metallic phase occurs at a critical concentration n_c. According to the scaling theory [6], the zero-temperature conductivity $\sigma(0)$ is expected to increase continuously for $n > n_c$. Nevertheless, in the expression of Mott [5] based on the Ioeffe–Regel [7] criterion and Anderson localization [8], $\sigma(0)$ increases continuously with n merely until a minimum value of conductivity (σ_{min}). Experimentally, it is impossible to reach zero-temperature conductivity $\sigma(0)$. Instead of the direct measurement of $\sigma(0)$, it can be experimentally determined from the extrapolation of the measured low-temperature conductivity to zero-temperature if the conductivity data obey the electron–electron interaction model [9]. The first pioneering experiments were performed in turn demonstrating that n_c can be evaluated by using experimental $\sigma(0)$ data as a function of the doping concentration just above the n_c in Si and Ge [10,11]. However, for $n \gg n_c$, the carrier concentration dependent conductivity is expressed in terms of Born conductivity (σ_{Born}) [12].

$In_xGa_{1-x}N$ alloys exhibit different electrical features, depending on the indium composition range. For instance, the mobility of $In_xGa_{1-x}N$ samples is expected to increase with the increase of x for $x > 0.9$, while it decreases with x for $x < 0.2$ [13].

In the present paper, in order to provide insight into the low-temperature behavior of $In_xGa_{1-x}N$ alloys, we have investigated

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the MIT in a series of $In_xGa_{1-x}N$ alloys as a function of the indium composition for $x \leqslant 0.135$.

2. Experimental

The InGaN epilayers that are presented in the present work were grown in an atmospheric pressure vertical MOVPE reactor with a showerhead configuration. Standard ammonia, TMGa and TMIn precursors were employed, while N_2 was always used as the main carrier gas. However, H_2 was also introduced in the reactor through two channels: first, it was always used as a carrier gas for the alkyls and, second, in controlled amounts via an additional upline. This allowed for an investigation into the role of the overall H_2 partial pressure in controlling the In incorporation. The TMGa precursor was delivered to the reactor via a double-dilution line, which allows for changing the Ga molar fraction while maintaining constant hydrogen flow that was injected into the growth chamber. The TMIn was instead introduced via a standard single-dilution line, where a given hydrogen flow entered into the bubbler and dragged a quantity of TMIn depending on the temperature and bubbler pressure, which were controlled, respectively, by a thermostatic bath and an online pressure controller. Obviously with this system, the TMIn molar fraction delivered to the growth chamber was always proportional to the hydrogen flow. The 2″ sapphire substrates were rotated around their axis at rates varying between 120 and 750 rpm: the change in rotation speed enabled the controlling of the growth rate, which had a strong influence on the indium content in the alloy as reported previously [14,15]. 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 approximately 7000), and an InGaN alloy deposited at 800 °C with different conditions, as reported above, in order to vary the In content.

The high-resolution X-ray diffraction (HRXRD) measurements were performed by a D8/Bruker diffractometer, which was equipped with a Cu source and a Ge(0 2 2) monochromator. The In composition was determined by HRXRD assuming that the lattice parameter varies linearly with the In fraction according to Vegard's law. The obtained In composition values are in excellent agreement as reported previously [14]. The thickness of the InGaN epilayers was approximately 600 nm with the indium composition varying from 0.060 to 0.135.

For the resistivity and Hall effect measurements by the van der Pauw method, square shaped (5×5 mm^2) samples were prepared with four contacts in the corners. By using annealed indium dots, the ohmic contacts to the samples were prepared and their ohmic behavior was confirmed by the current–voltage characteristics. Measurements were made at temperature steps over a temperature range of 15–320 K using a Lake Shore Hall effect measurement system (HMS) [3]. At each temperature step, the Hall coefficient (with a maximum 5% error) and resistivity (with a maximum 0.2% error in the studied range) were measured for both current directions. The magnitude of the magnetic field was 0.5 T.

3. Results and discussion

Fig. 1 shows the activation energy (ΔE_a) as a function of the indium concentration x, deduced from the temperature dependent conductivity data of the $In_xGa_{1-x}N$ alloys ($0.06 \leqslant x \leqslant 0.135$) [3] using the Arrhenius relation $\sigma(T) = \sigma_a \exp(-\Delta E_a/k_B T)$. Here, k_B is Boltzmann's constant and σ_a is a parameter depending on the semiconductor nature. $In_xGa_{1-x}N$ samples show semiconductor-like behavior, and the conductivity increases as the temperature increases [3]. However, the increase in conductivity with the temperature is very low when the indium composition (x) drops to a value of 0.06. The results of the conductivity measurements apparently indicate $\Delta E_a \to 0$ in the $In_xGa_{1-x}N$ system with a further reduction in x.

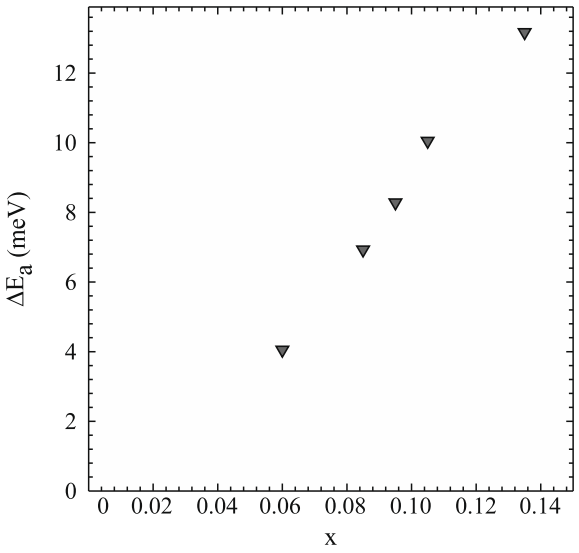

Fig. 1. Indium composition (x) dependence of the thermal activation energy plotted as ΔE_a vs. x.

The Bohr radius is given with relation $a_B^* = 4\pi\varepsilon_0\varepsilon\hbar^2/m^*e^2$, where ε_0 is the permittivity of vacuum, $\hbar$ is Planck's constant and e is the electron charge. By using an iterational method [13], the values of a_B^* of $In_xGa_{1-x}N$ alloys as a function of x can be calculated with the values of effective masses $m^* = 0.22\ m_0$ and $0.115\ m_0$, and the static dielectric constants of $\varepsilon = 10.4$ and 15.3 for GaN and InN, respectively. The critical density for the metal–insulator transition is obtained by using the relation $n_c = (0.25/a_B^*)^3$ as a function of x [3]. Our $In_xGa_{1-x}N$ alloys with $n > n_c$ fall on the metallic side of the MIT. In case of $n < n_c$, the system will be on the insulating side of the MIT.

In a previous work [16], we showed a high bowing parameter ~3.6 eV in $In_xGa_{1-x}N$ layers, which indicates the presence of disorder in the structure. The prediction of Mott is that every disordered material must pass a minimum metallic conductivity (σ_{min}) that is given by

$$\sigma_{min} = C\left(\frac{e^2}{a\hbar}\right), \tag{1}$$

where a is the distance between the centers, which equals approximately $n_C^{-1/3}$. C is a numerical constant at an order of 0.03 [5]. However, the values of $C = 0.06$ and 0.12 were predicted in p-type and n-type materials, respectively [9]. The calculated values of the σ_{min} for $C = 0.03$ are shown in Table 1.

We characterize the transport properties of the $In_xGa_{1-x}N$ samples in terms of the resistivity ratio, $\rho_r = \rho\ (15\ K)/\rho\ (300\ K)$. The temperature dependence $\sigma(T)$ of the investigated samples is very weak with $\rho_r = 1.11$–1.75. One can expect that the value of ρ_r becomes very high on insulating side of the MIT. We conclude, therefore, that the investigated samples are on the metallic side of the MIT. $\sigma(0)$ is finite, but the sign of the temperature coefficient of resistivity ($d\rho/dT$) remains negative for the investigated samples, as in disordered metals. Normally, $d\rho/dT > 0$ indicates metallic state, but $d\rho/dT < 0$ corresponds to insulating state. In our case, the observed features suggest that the three-dimensional (3D) localization-interaction model for disordered metallic systems above the MIT can be used for an explanation of low-temperature metallic transport in $In_xGa_{1-x}N$ samples [17–19]. According to this

Table 1
Electrical parameters of $In_xGa_{1-x}N$ alloys.

x	ρ_r	l (Å)	$k_F\, l$	$\sigma(0)$ $(\Omega\,cm)^{-1}$	σ_{min} $(\Omega\,cm)^{-1}$	λ_F (Å)
0.06	1.11	23.2	2.19	169.7	6.87	64.7
0.085	1.46	5.43	0.329	16.35	6.71	103.7
0.095	1.57	4.65	0.287	13.08	6.65	101.9
0.105	1.73	2.56	0.139	6.411	6.58	115.4
0.135	1.75	2.42	0.121	4.99	6.41	125.2

model, the conductivity of 3D disordered metallic systems is written as [19]

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

where the $mT^{1/2}$ term arises from electron–electron interactions. The sign of m is positive for the investigated samples [3]. The negative sign of m can be only found for the more metallic samples with $d\rho/dT > 0$.

It is not easy to distinguish whether the 3D or the two-dimensional (2D) limit is the appropriate in a disordered system. In the 2D structures, the electron–electron interaction term ($mT^{1/2}$) depends logarithmically on temperature. In the 2D structures, electrons from donors are distributed among the available states in the valleys located at the outskirts of the Brillouin zone [20]. The 2D layer is metallic when the valleys are equally populated or spin polarization is absent, whereas it becomes insulating when valley and spin polarizations are sufficiently large [20]. In a previous work, we showed that the temperature dependent the conductivity of the $In_xGa_{1-x}N$ samples can be well explained by the 3D model that takes into account electron–electron interaction and weak localization with the parameters that are in agreement with the theoretical predictions. Therefore, we consider the MIT in 3D for the investigated samples. This model predicts that electron–electron interactions play an important role in the low-temperature transport, whereas weak localization effects are dominant at higher temperatures.

Fig. 2 shows the calculated conductivity values by using various theoretical conduction model and the experimental values of zero-temperature conductivity ($\sigma(0)$) as a function of x. Since the carrier concentration $In_xGa_{1-x}N$ layers are nearly temperature-independent, we can accept the same value of the carrier concentration for the all temperature points even at $T \to 0$ in our theoretical calculations [3]. The values of $\sigma(0)$ can be experimentally determined by using Eq. (2). When the Fermi wavelength λ_F is much smaller than the mean free path l of the carriers, the Boltzmann approach can successfully describe the transport properties of normal metals. For $In_xGa_{1-x}N$ alloys which are in metallic side of MIT, as the x is increased, the structural or composition disorders are increased, the mean free path (l) becomes small, and eventually it may become smaller than λ_F. As can be seen in Table 1, $\lambda_F > l$ for our $In_xGa_{1-x}N$ alloys. In this case, some correction terms are added to the low-temperature conductivity of $In_xGa_{1-x}N$ alloys. Such a situation can be explained by the presence of electron–electron interactions in the system [7]. It has been shown that impurity band conduction dominates the electron transport of high degenerate $In_xGa_{1-x}N$ samples ($0.06 \leqslant x \leqslant 0.135$) in a temperature range of 15–350 K. The temperature dependent conductivity can be well explained by the model that takes into account electron–electron interactions and weak localization [3]. However, for low-temperature analysis, we use the values of $\sigma(0)$ determined for $T < 50$ K.

It can be seen in Table 1 that $\sigma(0) > \sigma_{min}$ for $x = 0.06$, 0.085 and 0.095, but $\sigma(0) < \sigma_{min}$ for $x = 0.105$ and 0.135. In the case of $\sigma(0) > \sigma_{min}$, the conductivity is given by the Kubo-Greenwood formula [5]

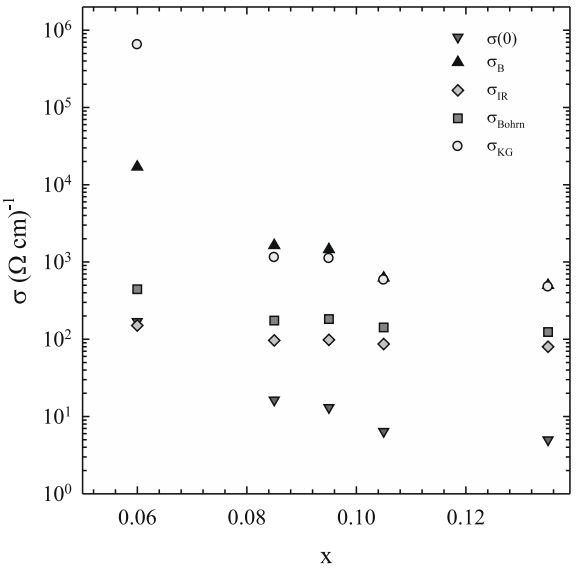

Fig. 2. Indium composition (x) dependence of the zero-temperature conductivity plotted as $\sigma(0)$ vs. x. Different symbols represent different values of the conductivities calculated by the various theoretical models (see text).

$$\sigma_{KG} = \sigma_B(1-R)^2, \tag{3}$$

where $R = 3/2(k_F l)^2$, k_F is the Fermi wave vector ($k_F = (3\pi^2 n)^{1/3}$), l is the mean free path ($l = \sigma\hbar k_F/ne^2$) and σ_B is the classical Boltzmann conductivity that is written as [5]

$$\sigma_B = \frac{1}{3\pi^2}\left(\frac{e^2}{\hbar}\right)k_F^2 l. \tag{4}$$

Both σ_B and σ_{KG} values are very high compared to the experimental conductivity values. This situation may explain the presence of a high carrier concentration ($n \gg n_c$), which may be due to a large number of nitrogen vacancies in InGaN alloys.

For $n \gg n_c$, Born introduced a formulation to calculate conductivity as a function of the carrier concentration [12]

$$\sigma_{Born} = \frac{e^2}{2\pi\hbar a_B^*}\frac{(k_{F_v}a_B^*)^3}{\ln[1+1/\gamma] - 1/(1+\gamma)}, \tag{5}$$

where $\gamma = d/\pi k_{F_v} a_B^*$, d is the number of valleys screening each electron and $k_{F_v} = (\pi^2 n/2)^{1/3}$. In our calculations, we assume $d = 1$ for simplicity. If d is accepted as greater than unity, the values of σ_{Born} will be higher than the values of $\sigma(0)$. Although the values of σ_{Born} undergo the approximate experimental values, the values of σ_{Born} are still higher than the experimental values. This may arise due to the strongly disordered structure of $In_xGa_{1-x}N$ alloys. As can be seen from Table 1, values of $k_F\, l$ were found to be smaller than unity for all but one of the samples ($x = 0.06$). $k_F\, l < 1$ arises their low conductivity related to their strongly disordered structures.

In the opposite regime $\sigma(0) < \sigma_{min}$, the conductivity behavior can be described in terms of the scaling theory of localization [6]. According to the scaling theory of localization for $n > n_c$, $\sigma(0)$ is evaluated as a function of parameter t that describes the degree of the disorder and interaction [6]

$$\sigma(T=0,t) = \sigma_0\left(\frac{t}{t_c} - 1\right)^{v}, \tag{6}$$

where σ_0 is the prefactor and v is the critical exponent. Here, t and t_c can be accepted as the arbitrary parameter and critical arbitrary parameter, respectively. These parameters can be composition, doping concentration, stress, etc. In several strongly disordered systems, $v = 0.5$–1.6 has been found [5,9,12,21]. The scaling theory may be applicable, if n is very close to n_c [6]. According to Shlimak

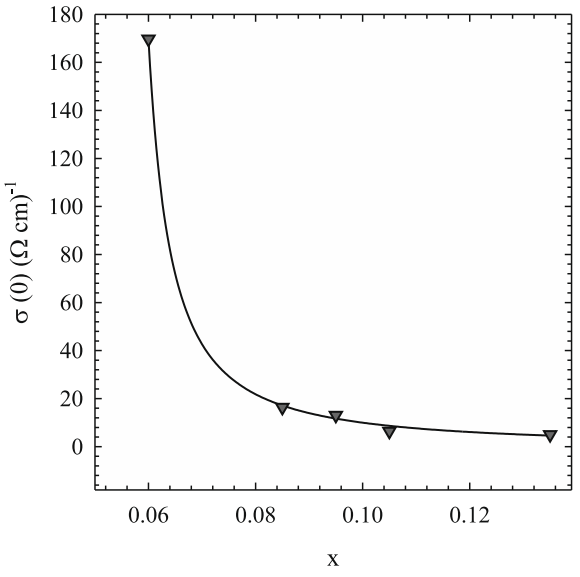

Fig. 3. Indium composition (x) dependence of the zero-temperature conductivity plotted as $\sigma(0)$ vs. x. The solid line is the best fit with Eq. (6) to the experimental data.

and Kaveh [9] $m = 0$ is the boundary between the scaling regime and metallic regime, and in this case the conductivity should be considered as the $\sigma_{\min}$. As the value of n is far from n_c, the sign of m will be negative [9]. However, for our $In_xGa_{1-x}N$ alloys, $d\sigma/dT$ is still greater than zero, i.e., $m > 0$. If one can also use $C = 0.12$ instead of $C = 0.03$ in Eq. (1), except for $x = 0.06$, the other samples will fulfill $\sigma(0) < \sigma_{\min}$ condition, which is necessary for the validity of the scaling theory. Both of the values of m are positive and the uncertainties in C lead to the result that the scaling theory may still be applicable for $In_xGa_{1-x}N$ alloys, if Ioeffe–Regel conductivity $\sigma_{IR} \geqslant \sigma(0)$ [5]. σ_{IR} is given as [7]

$$\sigma_{IR} = \left(\frac{2}{\pi^2}\right)\left(\frac{e^2}{ah}\right). \tag{7}$$

In Fig. 2, we calculated the values of the σ_{IR}, in which these values are close to the experimental value only for $x = 0.06$. Since the $k_F\, l \sim 2$ for $x = 0.06$, the condition of Ioeffe–Regel is fulfilled, while the other samples fall below the Ioeffe–Regel regime with $k_F\, l < 2$ (Table 1).

For the $In_xGa_{1-x}N$ alloys, we can estimate the critical indium composition (x_c) corresponding to MIT by using Eq. (6). Fig. 3 shows the $\sigma(0)$ vs. x. The solid line is the best fit to Eq. (6) with the values $\sigma_0 = 7.88 \pm 1.64\ (\Omega\,\mathrm{cm})^{-1}$, $x_c = (0.0543 \pm 6.8 \times 10^{-3})$, and $\nu = -1.36 \pm 0.682$. The negative sign of the ν arises due to the increments in conductivity with decreasing x in $In_xGa_{1-x}N$. $\nu > 1$ is observed in heavily compensated semiconductors [11]. The value of 1.36 for ν was observed in various strongly disordered systems [22–24]. In a previous work [3], low mobility values were observed in heavily compensated $In_xGa_{1-x}N$ alloys which exhibit disordered behavior. According to the scaling theory, $\sigma_0 > \sigma_{\min}$ is expected. This condition is fulfilled with the value of $\sigma_0 = 7.88$ for $In_xGa_{1-x}N$ alloys.

The value of 0.0543 for x_c would appear to be in reasonable agreement with the experimental observations. Actually, as can be seen in Fig. 1, a decrease in x can lead to temperature-independent behavior with zero activation energy. $In_xGa_{1-x}N$ alloys may completely behave like a metal for $x < x_c$ and one can expect $d\sigma/dT < 0$, i.e., conductivity decreases with increasing temperature.

4. Conclusion

We have shown that the indium composition (x) dependent conductivity of $In_xGa_{1-x}N$ alloys can be explained by the scaling theory along with the parameters that are in agreement with the theoretical predictions. The indium composition (x) dependent conductivity cannot be explained by the classical expressions for $In_xGa_{1-x}N$ alloys due to their strongly disordered compositions. Thermal activation energy decreases with the decrease in x. This shows that conduction moves to exhibit fully metallic behavior, which can be favorable for $x_c = 0.0543$.

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