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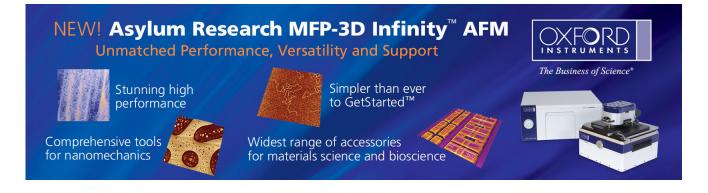
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Effects of alloy disorder on the transport properties of $AI_xGa_{1-x}N$ epilayers probed by persistent photoconductivity

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The effects of alloy fluctuations on the transport properties of Al_xGa_{1-x}N alloys ($x \sim 0.35$) have been probed through the use of persistent photoconductivity (PPC). In the PPC state, the electron mobility, μ_e , as a function of electron concentration, n, in a single sample can be obtained under controlled light illumination conditions. It was found that μ_e is a constant when n is below a critical value n_c and it then increases with n at $n > n_c$. This mobility behavior was attributed to the effects of alloy fluctuations in $Al_xGa_{1-x}N$ alloys. As a result, the initial PPC buildup kinetics seen in $Al_xGa_{1-x}N$ alloys was quite different from those in better understood semiconductor alloys, such as AlGaAs and ZnCdSe, and is a direct consequence of the observed unique dependence of μ_e on n. From these measurements, the total density of the tail states below the mobility edge in the conduction band was estimated to be 1.46×10^{17} cm⁻³ in a Al_{0.35}Ga_{0.65}N sample. The results were compared with those in II-VI semiconductor alloys and their implications on III-nitride device applications were discussed. © 2000 American Institute of Physics. [S0003-6951(00)02513-4]

The recent successful development of bright blue lightemitting diodes (LEDs) and laser diodes (LDs) has stimulated great interest in III-nitride materials, including GaN, $In_rGa_{1-r}N$, and $Al_rGa_{1-r}N$ alloys. Among them Al_rGa_{1-r}N alloys play a crucial role in many optoelectronic devices including blue/ultraviolet (UV) light emitters, UV detectors, and high-temperature/high-power transistors.^{1,2} However, the optical and electrical properties of $Al_rGa_{1-r}N$ alloys have not been well studied, especially for high Alcontent (x > 0.25) materials. It is expected that alloy fluctuations will play an important role in determining the optical and transport properties of $Al_xGa_{1-x}N$ alloys due to the large band gap difference between AlN (6.2 eV) and GaN (3.4 eV). However, studies on such effects in $Al_xGa_{1-x}N$ alloys have been scarce.

PPC is a light-induced enhancement in the conductivity that persists for a long period of time after the termination of light excitation and has been observed in many $III-V^{3-7}$ and II-VI^{8,9} semiconductors, also including p- and n-type GaN epilayers, AlGaN/GaN heterostructures, and AlGaN epilayers.^{10–18} According to the first-principal calculations,^{19,20} the PPC effect in III-nitride materials is a natural consequence of deep level impurities (DX or AX centers).

In this letter, we report the use of PPC to study the effects of alloy fluctuations on the electron transport properties of $Al_xGa_{1-x}N$ alloys. By utilizing the unique features of PPC, namely the very long lifetimes of the photoexcited carriers and the continued variation of the electron concentration in the conduction band, we were able to measure the electron mobility (μ_e) as a function of electron concentration (n) in a single sample. We found that μ_e is a constant when *n* is below a critical value (n_c) and it then increases with n at $n > n_c$. The initial PPC buildup kinetics in Al_{0.35}Ga_{0.65}N epilayers were found to be quite different from those in better understood semiconductor alloys, such as $Al_xGa_{1-x}As$ and $Zn_xCd_{1-x}Se$. By formulating the electron mobility as a function of concentration and the PPC buildup kinetics, we show that the feature exhibited in the initial PPC buildup is a direct consequence of the dependence of μ_e on n due to the effects of alloy fluctuations on the electron transport in the conduction band in $Al_xGa_{1-x}N$ alloys.

The undoped Al_{0.35}Ga_{0.65}N epilayers were grown by metalorganic chemical vapor deposition (MOCVD) on a *c*-plane sapphire substrate with a 200 Å GaN buffer layer. The growth temperature and pressure were 1060 °C and 150 Torr, respectively. The thickness of the $Al_xGa_{1-x}N$ epilayers was about 3400 Å. The Al content was about 35% as determined by x-ray diffraction and photoluminescence measurements. Ohmic contacts were prepared by directly bonding Al wires to the $Al_xGa_{1-x}N$ sample and the ohmic behavior was confirmed by the linear I-V characteristics up to 10 V. For PPC measurements, a 1.5 V bias was supplied to the sample, a Hg lamp with a main emission line at about 253.7 nm was used as an excitation light source, and an electrometer was used to monitor the current. The electron mobility was determined by Hall-effect measurements. To ensure that each set of data obtained under different conditions have the same initial conditions, the sample was kept in darkness for about 3 h to reach the true dark state before data acquisition. The photon flux was estimated to be about 3.0×10^{13} /cm² s at a maximum excitation light intensity, $I_{exc} = I_0$ and was varied by a set of neutral density filters. All results reported here were obtained at 300 K.

Figure 1 shows a typical PPC behavior for one of our $Al_{0.35}Ga_{0.65}N$ epilayers measured at $I_{exc}=I_0$. The timedependent PPC decay $I_{PPC}(t)$ can be very well described by a stretched-exponential function, which is frequently used to describe the PPC relaxation in a wide class of III-V and II-VI semiconductors7-11

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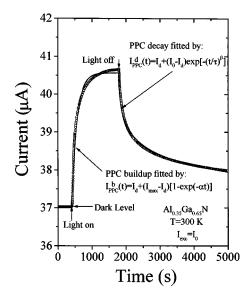


FIG. 1. Persistent photoconductivity (PPC) measured at 300 K for an $Al_{0.35}Ga_{0.65}N$ sample grown by MOCVD. The least squares fit for the PPC decay (open squares) with Eq. (1) and for the PPC buildup (open circles) with Eq. (2) is plotted as solid lines. The bold line near the saturation level indicates the disagreement between the experimental data and Eq. (2).

$$I_{\rm ppc}(t) = I_d + (I_1 - I_d) \exp[-(t/\tau)^{\beta}], \qquad (1)$$

where I_d is the initial dark current, I_1 is the current level immediately after the termination of the excitation source, τ is the decay time constant, and $\beta(0 \le \beta \le 1)$ is the decay exponent. The fitted values of τ and β for the data shown in Fig. 1 are 1350 s and 0.35, respectively.

The PPC buildup kinetics in AlGaAs has been measured and theoretically formulated in the context of DX centers to follow^{21,22}

$$I_{\rm PPC}^{b}(t) = I_d + (I_{\rm max} - I_d) [1 - \exp(-\alpha t)],$$
(2)

where α is a constant and I_{max} is the PPC saturation level. However, as illustrated in Fig. 1, the PPC buildup kinetics in Al_{0.35}Ga_{0.65}N epilayers studied here can no longer be described by Eq. (2) and in fact the use of Eq. (2) results in a poor fit, particularly near the PPC saturation region (bold part).

If the PPC buildup can be described by Eq. (2), then a linear time-dependent behavior at the initial PPC buildup stage, $I_{PPC}^b(t) \propto t(\alpha t \ll 1)$, is expected. We have carefully monitored the initial PPC buildup kinetics in Al_{0.35}Ga_{0.65}N epilayers by systematically varying I_{exc} . Figure 2 shows the time-dependent PPC buildup behaviors during the first 50 s measured for varying I_{exc} , which clearly illustrates that for the samples studied here, a linear time-dependent PPC buildup at the initial stage was not present.

On the other hand, we have shown in previous works that the electron transport properties in II–VI semiconductor alloys are strongly influenced by the tail states caused by the alloy disorder.^{23,24} In semiconductors (*n* type) with alloy fluctuations, the conductivity results mainly from the electron hopping between localized states when the electron quasi-Fermi level, E_F , is below the mobility edge, E_m .²⁵ Assuming that the Fermi distribution is a step function (at the electron flow temperatures) and that the alloy disorder induced an ex-

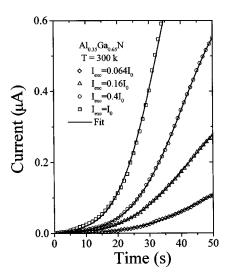


FIG. 2. The kinetics of the initial PPC buildup for an Al_{0.35}Ga_{0.65}N sample measured at four different excitation light intensities, i.e., $I_{exc} = I_0, 0.4I_0, 0.16I_0, 0.064I_0$. The solid curves are the least squares fit of data with Eq. (4). The dark currents have been subtracted out.

ponential tail state in the conduction band edge, a quadratic time-dependent initial PPC buildup was derived from the Kubo–Greenwood formula^{23,24}

$$I_{\text{PPC}}^{b}(t) = I_d + \text{const.}[1 - \exp(-\alpha t)]^2 \propto t^2 (\alpha t \ll 1), \quad (3)$$

which agreed well with experimental observations in $Zn_xCd_{1-x}Se$ alloys. The quadratic time-dependent initial PPC buildup behavior implies that the electron mobility $\langle \mu_e \rangle$ in the tail states of $Zn_xCd_{1-x}Se$ alloys is proportional to the electron concentration $\langle n \rangle$, $\langle \mu_e \rangle \propto \langle n \rangle$, since the conductivity $\sigma(t)$ is effectively proportional to the product of $\langle \mu_e n \rangle$, where $\langle \rangle$ stands for an assemble average and $\langle n \rangle$ is proportional to $[1 - \exp(-\alpha t)]^{.23,24}$

However, for $Al_{0.35}Ga_{0.65}N$ epilayers studied here, the initial PPC buildup behavior is neither linear nor quadratic with time. Instead, the observed PPC buildup behavior follows

$$I_{\text{PPC}}^{b}(t) = I_{d} + \text{const.}[1 - \exp(-\alpha t)]^{\gamma} \propto t^{\gamma}(\alpha t \ll 1), \quad (4)$$

where α and γ are constants. The least squares fits of the initial PPC buildup data with Eq. (4) are plotted in Fig. 2 as solid lines with the fitted value of γ being approximately 2.9±0.2 for different I_{exc} . We believe that the initial PPC buildup behavior in Al_{0.35}Ga_{0.65}N epilayers is also caused by the tail states in the conduction band induced by alloy fluctuations, similar to the case in ZnCdSe alloys. The feature exhibited by the initial time dependence seen in Al_xGa_{1-x}N alloys $(I_{\text{PPC}}^b(t) \propto t^{2.9 \pm 0.2})$ can be attributed to a unique functional dependence of $\langle \mu_e \rangle$ on $\langle n \rangle$ to be described below.

The experimentally measured functional form of $\langle \mu_e \rangle$ vs $\langle n \rangle$ in Al_xGa_{1-x}N alloys is shown in Fig. 3, which shows that μ_e is a constant when *n* is below a critical value n_c and it increases with *n* at $n > n_c$. This behavior is caused by the electron filling effects in the localized band tail states in AlGaN alloys. When $n < n_c$, electrons are localized in the tail states and contribute to the Hall mobility only through hopping or tunneling. As *n* increases and reaches a certain critical value n_c , the electron quasi-Fermi level, E_F , reaches above the mobility edge E_m , at which a transition from lo-

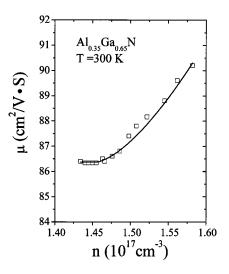


FIG. 3. The measured electron mobility as a function of electron concentration (open squares) and the least squares fit (solid line) of the data with Eq. (5).

calized electron transport to a free carrier transport takes place. Thus the electron mobility is expected to increase with *n* above n_c . The experimentally measured dependence of $\langle \mu_e \rangle$ on $\langle n \rangle$ in Al_xGa_{1-x}N alloys can be well described by

$$\mu_e = \mu_0 + A(n - n_c)^{\rho} \theta(n - n_c), \qquad (5)$$

where μ_0 is the electron mobility at $n < n_c$ and $\theta(n - n_c)$ is a step function, with A, n_c , and ρ being fitting parameters. The second term describes the conduction contributed by electrons with energies above E_m . The least squares fit of data with Eq. (5) is plotted as the solid line in Fig. 3 and the fitted values are $\mu_0 = 86.4 \text{ cm}^2/\text{V}$ s, $n_c = 1.46 \times 10^{17} \text{ cm}^{-3}$, and $\rho = 1.6$, respectively.

Since the measured electron concentration in darkness is about $1.43 \times 10^{17} \text{ cm}^{-3} \approx n_c$, so $(n-n_c)$ corresponds approximately to the photoexcited electron concentration, n_e . From the fitted value of $n_c (\approx 1.46 \times 10^{17} \text{ cm}^{-3})$, we can also conclude that the total density of the band tail states below the mobility edge in $\text{Al}_x \text{Ga}_{1-x} \text{N}$ alloys is about 1.46 $\times 10^{17} \text{ cm}^{-3}$, which is directly correlated with the degree of alloy fluctuations in these materials.

From Eq. (5) and Fig. 3, we have $\mu_e \propto [n_e(t)]^{1.6}(n > n_c)$. We therefore have $I_{PPC}^b(t) \propto \sigma(t) \propto \langle \mu_e n_e \rangle \propto [n_e(t)]^{2.6} \propto t^{2.6}$, since $n_e(t) \propto t$ at $\alpha t \ll 1.^{23,24}$ On the other hand, when we fit the initial PPC buildup kinetics shown Fig. 2 directly to Eq. (4), $I_{PPC}^b(t) \propto t^{\gamma}$, we obtain $\gamma = 2.9 \pm 0.2(\alpha t \ll 1)$, which is very close to the value obtained from the mobility measurements. Thus we can conclude that the initial PPC buildup kinetics shown in Fig. 2 is a natural consequence of the unique relationship between μ_e and *n* described by Eq. (5).

Different relationships between μ_e and *n* as well as the initial PPC buildup kinetics observed in Al_xGa_{1-x}N and Zn_xCd_{1-x}Se alloys may be due to the fact that the band gap difference between AlN and GaN ($\Delta E_g = 2.8 \text{ eV}$) is much larger than the difference between ZnSe and CdSe ($\Delta E_g = 1.1 \text{ eV}$), which may result in a stronger effect of alloy fluctuations as well as a different distribution of the tail states in Al_xGa_{1-x}N alloys.

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alloys have been investigated. Through the use of PPC, we found that the electron mobility is a constant of about 86.4 cm²/V s below the mobility edge. As the electron concentration *n* increases and reaches a certain critical value n_c , the electron mobility increases with *n* following μ_e $\propto [n_e(t)]^{1.6}(n > n_c)$. From this unique dependence of μ_e on n_e , we thus expect the time dependence of the initial PPC buildup to follow $I_{\rm PPC}^{b}(t) \propto t^{\gamma}$ with $\gamma \approx 2.6$, which is consistent with $\gamma \approx 2.9$ observed experimentally. By measuring the electron mobility as a function of the electron concentration in the PPC state, the total density of the conduction band tail states below the mobility edge in Al_{0.35}Ga_{0.65}N epilayers was also estimated to be around $1.46 \times 10^{17} \, \text{cm}^{-3}$. Our results have demonstrated that alloy disorder strongly influences the transport properties of AlGaN alloys and that the PPC effect can be utilized to probe the properties induced by alloy fluctuations, such as the density of the tail states and the mobility behavior near the mobility edge. Moreover, our results presented here indicate that for III-nitride device applications using $Al_xGa_{1-x}N$, effects of alloy fluctuations are important even at room temperature due to the large band gap difference between GaN and AlN.

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In summary, the effects of tail states induced by alloy disorder on the electron transport properties in $Al_xGa_{1-x}N$