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Temperature dependence of carrier lifetimes in InN

Fei Chen[*, 1], **A. N. Cartwright**[1], **H. Lu**[2], and **W. J. Schaff**[2]

[1] Department of Electrical Engineering, University at Buffalo, State University of New York, Buffalo, NY 14260, USA

[2] Department of Electrical and Computer Engineering, Cornell University, Ithaca, New York 14853, USA

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Time-resolved pump–probe transmission measurements were used to determine the temperature dependence of carrier lifetime for InN epilayers with unintentionally doped levels from 10^{18} to 10^{19} cm^{-3}. The observed decay time at 20 K is well explained by a dominating radiative interband recombination, while at room temperature it is attributed to a defect related nonradiative recombination channel. The temperature dependence of the radiative lifetime is deduced from the measurements of both differential transmission decay time and PL intensity. For the best quality sample, we find the radiative lifetime increases proportionally to $T^{3/2}$, as theory predicts when a *k*-selection rule holds, which suggests that the radiative band-to-band transition accounts for the observed infrared photoluminescence over the entire temperature.

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1 Introduction

The recent achievements of optical characterization of high quality wurtzite InN crystals, although still a subject of debate, have revised the band gap of InN to around 0.7 eV [1, 2], much narrower than the ~1.9 eV that was measured in earlier studies [3]. This unexpected discovery has provided new opportunities for potential applications of InN based optoelectronic devices, such as infrared emitters and solar cells. However, many questions remain open concerning the mechanisms that rule carrier recombination and lifetimes in these materials. The understanding of the physics of recombination mechanisms can be of significant help for revealing the true band gap of InN and for the development of InN based devices. In this paper, we investigated the carrier recombination dynamics and determined the carrier lifetime in InN from 20 K to room temperature by employing time-resolved pump–probe transmission technique. The measured carrier lifetimes at 20 K are roughly consistent with the calculated bimolecular radiative lifetimes considering a band-to-band free carrier transition. Temperature dependence of the derived radiative lifetime in the best quality sample shows an increase proportional to $T^{3/2}$, as theory predicts, indicating that the origin of the infrared photoluminescence (PL) is indeed dominated by this interband recombination.

2 Experiments and results

Three unintentionally doped InN films were grown on (0001) sapphire with different buffer layers by molecular beam epitaxy. The details of the growth technique have been reported elsewhere [4]. The structures contain (i) sample A, GaN(220 nm)/InN(850 nm); (ii) sample B, AlN(300 nm)/InN(350 nm);

[*] Corresponding author: e-mail: feichen@acsu.buffalo.edu, Phone: +01 716 645 3123

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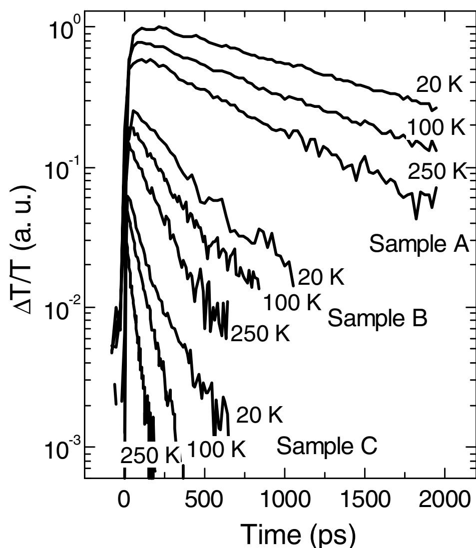

Fig. 1 Differential transmission transients of sample A, B, and C at different temperatures as indicated in the figure, under the pump fluence of 1 μJ/cm^2.

and (iii) sample C, GaN(220 nm)/InN(600 nm). The mobility and free electron concentration were measured at room temperature as 1340 cm^2/Vs and 1.3×10^{18} cm^{-3} for sample A, 820 cm^2/Vs and 2.7×10^{18} cm^{-3} for sample B, 826 cm^2/Vs and 1.2×10^{19} cm^{-3} for sample C, respectively. It has been reported that there is only a slight change of free electron concentration and mobility over the temperature range from 10 K to 300 K for an InN sample grown under the same growth technique [5]. Hence, in this work a weak temperature dependence of mobility and free electron concentration is assumed on all three samples.

Time-resolved differential transmission (pump–probe transmission) measurements were performed using a Coherent regenerative amplifier (REGA), operating at 250 kHz. The output of the REGA was frequency doubled to 400 nm to serve as the pump source for an optical parametric amplifier that generates a tunable infrared pulse (0.9 μm to 3.6 μm) with typical pulse widths of ~300 fs. In these experiments, the tunable infrared pulse was used as the probe beam and the remaining output of REGA at 400 nm served as the pump beam. Continuous-wave PL spectroscopy was also performed on these samples by using an Ar$^+$ laser with a photon energy of 3.41 eV. The samples were cooled to approximately 20 K using a closed cycle helium refrigerator. The detailed temperature dependence of PL and absorption has been reported earlier, where a strong emission around 0.7 eV was observed [6, 7].

Figure 1 shows the differential transmission transients, with the probe energy corresponding to the absorption edge, at different temperature under a low fluence of 1 μJ/cm^2. The maximum injected carrier density achieved here is ~2.5×10^{16} cm^{-3}, much less than the free electron concentration in each sample. It is interesting to note that most of these transients can be described by a single exponential decay, and the carrier lifetime is extracted by the decay constant. As shown in Fig. 2, we observed that the carrier lifetimes decreased from 670 ps to 408 ps for sample A, 383 ps to 153 ps for sample B, and 98 ps to 43 ps for sample C, with increasing temperature from 20 K to 300 K. For the samples investigated here, our results demonstrate that carrier lifetime is inversely proportional to the free electron concentration both at room temperature and at low temperature. Thus, a linear curve can be obtained when plotting the carrier lifetime versus free electron concentration on a log–log scale (Fig. 2). A significant reduction in carrier lifetime due to the increase of free electron concentration is obvious here.

There are three main channels for the photogenerated carriers to recombine: nonradiative defect related, bimolecular radiative interband, and nonradiative Auger recombination. Under a low injection level, the carrier lifetime can be described by $1/\tau = 1/\tau_{\text{defect}} + B_r n_0 + B_{\text{Auger}} n_0^2$, where B_r and B_{Auger} are interband and Auger recombination coefficients, τ_{defect} is the defected related nonradiative lifetime, and n_0 represents the concentration of majority carriers (free electrons in our case). Remember that τ_{defect} is re-

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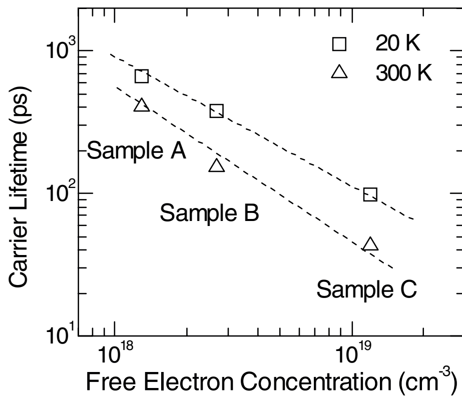

Fig. 2 Measured carrier lifetimes for sample A, B, and C at 20 K and 300 K.

lated to the defect density N_{defect} by the following expression: $1/\tau_{defect} = \sigma \upsilon N_{defect}$, where σ is the carrier capture cross section and υ represents the carrier velocity. The above equations imply that, for radiative interband and nonradiative Auger recombination, the carrier lifetime is inversely proportional to n_0 and n_0^2 respectively, while the defect related non-radiative recombination rate is proportional to the defect density. Hence, the inverse proportionality between n_0 and τ observed in the InN samples studied here, rules out the possibility of Auger recombination.

Our previous work suggested that the measured carrier lifetime at room temperature for these samples is consistent with trapping of the photogenerated carriers by impurities and defects, resulting in the fast nonradiative recombination, because the donor-like defects or impurities may also stimulate the formation of (or work as) the nonradiative recombination centers, reducing the carrier lifetime [6, 7]. In order to elucidate the recombination nature at low temperature, we calculated the bimolecular radiative recombination coefficient B_r, by using the method developed by other groups [8, 9]. The B_r is determined as $(2.6 \pm 0.5) \times 10^{-11}$ cm^3 s^{-1} at 300 K and $(1.5 \pm 0.3) \times 10^{-9}$ cm^3 s^{-1} at 20 K, respectively. Together with the measured free electron concentration in each sample, the calculated radiative lifetimes are then obtained by: $(B_r \times n_0)^{-1}$, and are listed in Table 1. We observed that the measured carrier lifetime is significantly shorter than the calculated radiative lifetime at 300 K, as a result of the dominating nonradiative recombination channel. Conversely, at 20 K the measured lifetime is consistent with the calculated radiative lifetime. The slight underestimate may be due to the uncertainty of the electronic parameters of InN (such as: effective hole and electron masses) used in the calculation. Furthermore, since the carrier densities come from Hall measurements interpreted with a single layer approximation, the measured electron concentration could be higher than that of the bulk InN layer due to a surface and interface electron accumulation [10–12]. Nevertheless, our results suggest the carrier recombination of these samples at 20 K is in good agreement with the radiative band-to-band transition.

In order to get further information on the temperature dependence of radiative lifetime, we can make the reasonable assumption that the variation of PL intensity versus temperature $I_{PL}(T) = \eta(T) I_{PL}(0)$ is the same as that of the internal radiative efficiency $\eta(T) = \tau_{nr}/(\tau_{nr} + \tau_r)$ [13]. Here, τ_r and τ_{nr} are the radiative and nonradiative lifetimes, and the measured carrier lifetime is simply given by $1/\tau = 1/\tau_{nr} + 1/\tau_r$. Thus, we can determine the radiative lifetime as $\tau_r(T) = \tau(T)/\eta(T) = \tau(T) I_{PL}(0)/I_{PL}(T)$, based on the measurements of both differential transmission decay time and PL intensity.

Table 1 Comparison of the measured carrier lifetime and the calculated radiative lifetime.

temperature	sample A	sample B	sample C
measured carrier lifetime at 20 K	670 ps	383 ps	98 ps
calculated radiative lifetime at 20 K	513 ps	333 ps	56 ps
measured carrier lifetime at 300 K	408 ps	153 ps	43 ps
calculated radiative lifetime at 300 K	29 ns	13 ns	3.2 ns

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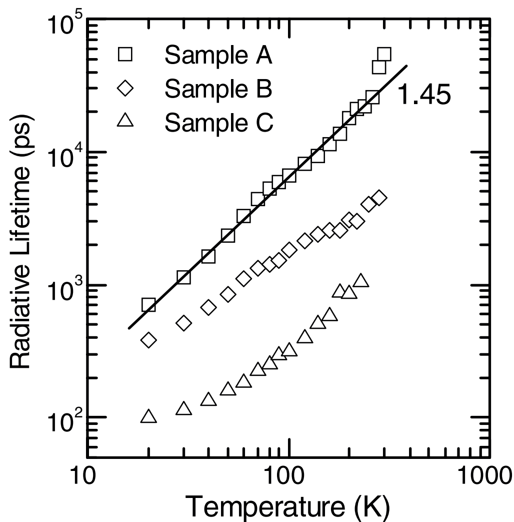

Fig. 3 Double logarithmic plot of the derived radiative lifetime in sample A, B and C as a function of temperature. The solid line indicates a $T^{1.45}$ fit for sample A.

Since nonradiative channels at the lowest temperature are considered to be non-active ($\eta = 1$ at 20 K), the temperature dependence of the radiative lifetime for each sample can be calculated qualitatively and is plotted in Fig. 3 on a double logarithmic scale. Because of a weak temperature dependence of the free electron concentration, the main temperature dependence of the radiative lifetime is only related to the bimolecular recombination coefficient B_r. For direct band gap semiconductors, B_r is proportional to $T^{-3/2}$ when *k*-selection rule holds [14], and the resulting radiative lifetimes are proportional to the $T^{3/2}$. As theory predicts, the radiative lifetimes for sample A give a $T^{1.45}$ dependence over almost the entire temperature range. This temperature behaviour indicates that the radiative band-to-band transition indeed governs the observed infrared luminescence. In contrast, there is a discrepancy between theory and experimentally determined temperature dependence for sample B and sample C. Let us remember that in InN the Fermi surface in the conduction band has shown a strong dependence on the free electron concentration due to the small effective electron mass [15]. The high free electron concentrations for sample B and sample C could raise the Fermi level into the conduction band, and cause a collapse of the *k*-selection rule in such highly degenerate cases. Moreover, the temperature dependence of reabsorption factor, absorption coefficient, refractive indices, and band gap shrinkage could also complicate our understanding.

3 Conclusions

In conclusion, we have studied the temperature dependence of the carrier lifetime in a series of InN epilayers, with varying unintentionally doping levels. We have found the observed decay time at 20 K is consistent with the radiative band-to-band recombination mechanism. For the best quality sample, the derived radiative lifetime increases proportionally to $T^{3/2}$ as theory predicts when a *k*-selection rule holds.

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