Ultrafast carrier thermalization in InN

Yu-Chieh Wen and Cheng-Ying Chen
Department of Electrical Engineering, National Taiwan University, Taipei 10617, Taiwan, and Graduate Institute of Electro-Optical Engineering, National Taiwan University, Taipei 10617, Taiwan

Chang-Hong Shen and Shangjr Gwo
Department of Physics, National Tsing-Hua University, Hsinchu 30013, Taiwan

Chi-Kuang Sun
Department of Electrical Engineering, National Taiwan University, Taipei 10617, Taiwan, and Graduate Institute of Electro-Optical Engineering, National Taiwan University, Taipei 10617, Taiwan

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Carrier thermalization dynamics in heavily doped n-type InN was investigated at room temperature with a femtosecond transient transmission measurement. The dependence of hot carrier cooling time on the total electron density indicates that the plasma screening of electron-LO phonon interactions is responsible for the reduction in energy-loss rate. Under low excitation, the carriers with different excess energies can be found to cool down with a fixed thermalization time of 1.4 ps. Intensity dependent study suggests that this relatively slow carrier cooling time could be attributed to the screening effect by high-background-doping plasma rather than the hot phonon effect. © 2006 American Institute of Physics. [DOI: 10.1063/1.2402899]

With a recent discovered narrow badgap, indium nitride (InN) has attracted attention for its potential applications in optoelectronics. The excellent electron transport properties of InN, including high mobility and high saturated drift velocity, also allow it to be developed as electronic devices with superior performances. To realize the various device applications, it is important to understand the carrier relaxation processes in InN.

InN is a highly polarized material with a high LO phonon energy. Strong carrier-polar optical phonon interactions are expected and should result in a sub-100 fs electron-LO phonon scattering time, according to theoretical predictions. One would expect that hot carriers could efficiently release its energy to the lattice on the order of or less than 100 fs. Experimentally, Zanato et al. had recently reported an electron-LO phonon scattering time of ∼200 fs by a frequency domain mobility measurement. However, in a recent time-resolved study with electron temperature carefully measured, Chen et al. discovered a prolonged hot carrier cooling with a time constant much longer than 1 ps even up to 10 ps. Ascáubi et al. also reported the observation of a 2–3 ps carrier cooling time under high photoexcitation, independently verified also by another absorption bleaching effect study. These time-domain data indicated that the observed carrier relaxation rate is much slower than previous theoretical expectation and were interpreted as a possible result affected by the hot phonon effect. In many material systems, the hot phonon effect is responsible for the reduction of energy-loss rate via Fröhlich interaction under high photoexcitation due to reabsorption of nonequilibrium phonons, with a characteristic cooling time prolonged to the phonon lifetime. However, a recent Raman measurement in high quality InN samples indicated a room-temperature (RT) LO phonon lifetime of 0.7 ps. With observed cooling time much longer than the RT LO phonon lifetime, these previous studies imply that besides the hot phonon effect, there could be other mechanisms responsible for the reduction of energy-loss rate in these studied InN samples.

In this letter, we report femtosecond time-resolved investigation on ultrafast carrier dynamics in a wurtzite InN epilayer, with high background n-type doping, at RT. The dependence of the observed carrier thermalization time on total electron density indicates that the electron-plasma screening of electron-LO phonon interaction plays an important role on the reduction in energy-loss rate. When we reduced the optical excitation to disturb the background electron Fermi sea in the perturbative regime, an intensity study suggests that the observed relatively slow carrier cooling time of 1.4 ps could be attributed to the screening effect by high-background-doping plasma rather than the hot phonon effect.

Degenerate pump-probe transient transmission measurements were performed using a femtosecond optical parametric oscillator with a 76 MHz repetition rate, tunable from 1240–1600 nm (1–0.77 eV) with ∼200 fs pulse-widths. Two lenses with different focal lengths were used to focus beams into the sample for a large excitation range. The radii of the focused pump and probe beams were kept the same and measured to be ∼7.5 and ∼39 μm for two adopted lenses. The pump fluence was tuned between 0.06 and 60 μJ/cm².

The sample under study was a wurtzite InN epitaxial film grown by plasma-assisted molecular beam epitaxy. By using a double-buffer layer technique, a high-crystalline-quality InN film was grown on a Si(111) substrate. The film thicknesses are 300 nm for the InN epilayer and 30 nm for the AlN buffer layer. With unintentional doping, a high-background free-electron concentration of 6 × 10¹⁸ cm⁻³ was determined by the Hall measurement at RT. Due to the existence of dense electrons, the states near the bottom of the conduction band are occupied, which results in an interband absorption edge energy much higher than the actual badgap energy. The RT photoluminescence peak was measured around 0.65 eV, while the optical absorption spectrum indicated an absorption edge of ∼0.8 eV. To distinguish the studied carrier cooling process from carrier recombination
process, which strongly depends on the doping density and sample quality and could be with a lifetime also of a few picoseconds, we performed transient transmission measurement with a photon energy close to the absorption edge of the studied sample. The inset of Fig. 1 shows a measurement result with 0.925 eV photon energy and a photocarrier density of $8 \times 10^{16}$ cm$^{-3}$. Our study indicated a carrier recombination time much longer than 30 ps, even under high photoinjection condition.

Figure 1 shows the normalized optical transient transmission changes measured with 1300 nm (0.953 eV) excitation wavelength upon different pump fluences ranging from 0.11 to 30 $\mu$J/cm$^2$, corresponding to effective photocarrier densities of $1.7 \times 10^{16}$–$4.6 \times 10^{18}$ cm$^{-3}$. According to the study by Chen et al., photoinjected carriers rapidly thermalize with background doping carriers after optical excitation. The whole carrier distribution then cools down with a definable carrier temperature with a time constant on the order of a few picoseconds. The photocarriers will eventually relax through carrier recombination. To quantitatively estimate the influence of photocarrier density on the hot carrier cooling, a simple convolution fitting was performed for the data in Fig. 1 with an impulse response function of the form $u(t)[a_1 e^{-t/\tau_1} + a_2 e^{-t/\tau_2}]$, where $u(t)$ is a unit step function, $a_1$ term is a faster exponential decay component with a time constant $\tau_1$ representing the carrier cooling process, and $a_2$ term is a slower exponential decay component representing the carrier recombination process. Through the fitting procedure, the interested carrier cooling time $\tau_1$ can be extracted and found to increase from 1.44(±0.16) up to 4.4(±1.3) ps. Our transient transmission study thus agrees with previous time-domain InN studies observing a prolonged carrier cooling time and supports a carrier-density dependent carrier cooling process. With the recent Raman measurement indicating a 0.7 ps RT LO phonon lifetime in InN, which is much shorter than our observed hot carrier cooling time even under low fluence excitation, the hot phonon effect should not be responsible for our observed reduction in energy-loss rate.

Screening of electron-LO phonon interaction by dense electron plasma could be a possible mechanism for the reduction in energy-loss rate in heavily doped InN. With an increase in the carrier density, the screening of the LO phonon emission becomes strong and makes a significant contribution when the density is up to the critical density, corresponding to the resonance density of LO phonons with plasmons. Due to the native $n$-type character of InN, the background doping density is usually comparable to or higher than this critical density, which is $\sim 1.5 \times 10^{18}$ cm$^{-3}$. This screening effect could thus seriously affect the energy-loss rate and could be responsible for the prolonged thermalization process observed up to date.

According to a theoretical study on the screening effect, the density dependence of phonon emission frequency via the screened Fröhlich interaction is proportional to $(1 + (N/N_F)^{2/3})^{-1}$, where $N$ and $N_F$ are carrier density and critical carrier density, respectively. This static-screening-approximation-based model allows one to investigate the influence of density on the screening effect quantitatively, although it would overestimate the reduction in LO phonon emission rate when the plasmon frequency $\omega_{pl}$ is close to the LO phonon frequency $\omega_{LO}$ due to dynamic Coulomb interactions. By using the electron density at which $\omega_{pl} = \omega_{LO}$ as the critical density, the normalized LO phonon emission rate can be estimated and is shown as a solid line in Fig. 2. This theoretical curve is normalized to the unscreened emission rate at a low carrier density. Here, the electron plasmon frequency is expressed as $\omega_{pl}^2 = (4 \pi e^2 N)/(\varepsilon_s m_e)$, where $\varepsilon_s$ is the high-frequency dielectric constant and $m_e$ is the effective electron mass. Physical parameters, $\varepsilon_s = 6.7$, $m_e = 0.07m_0$, and $\omega_{LO} = 590$ cm$^{-1}$, are used in the calculation. Reduction of LO phonon emission rate with a factor $>10$ can be found. Figure 2 also shows the inverse of the observed hot carrier cooling time $(1/\tau_1)$ versus total electron density, extracted from Fig. 1. It should be noted that the influence of the screening effect is associated with carrier temperature. To study the density dependent results under a similar temperature condition, we analyzed the data with photocarrier densities less than $5 \times 10^{18}$ cm$^{-3}$.

FIG. 1. (Color online) Normalized transient transmission change as a function of time delay measured with a photon energy of 0.953 eV and photocarrier densities of (from $a$ to $a'$) $5 \times 10^{16}$, $2 \times 10^{16}$, $9 \times 10^{15}$, $1.4 \times 10^{16}$, $7 \times 10^{16}$ (black lines), and $1.7 \times 10^{16}$ cm$^{-3}$ (red line). The inset shows the transient transmission change measured with a photon energy of 0.925 eV and a photocarrier density of $8 \times 10^{16}$ cm$^{-3}$. The steplike relaxation indicated $a > 100$ ps carrier lifetime under low excitation.

FIG. 2. Normalized LO phonon emission rate (solid line) as calculated following the description in the paragraph and the inverse of the carrier cooling time (open circles), which was measured with a photon energy of 0.953 eV, with respect to the total electron density $N$. The dash line shows the scattering strength of the phononlike branch of the plasmon-LO phonon coupled modes at long-wavelength limit as given by the theory in Ref. 24. Deviation between two theoretical curves is much less than our experimental uncertainty, suggesting that both models can describe our observed phenomenon well.
In summary, ultrafast carrier thermalization in InN was investigated at room temperature. The dependence of hot carrier cooling time on the total electron density indicates that the screening of electron-LO phonon interaction could be responsible for the significant reduction in energy-loss rate observed in InN. When disturbing the background Fermi sea in the perturbative regime, the carriers can be found to cool down with a thermalization time of 1.4 ps. This slow cooling rate was also suggested to be attributed to the screening effect rather than the hot phonon effect.

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