Subpicosecond time-resolved Raman studies of electron–longitudinal optical phonon interactions in InN

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Electron–longitudinal optical phonon scattering rate in InN has been directly measured by subpicosecond time-resolved Raman spectroscopy. The experimental results show that for a thick layer of InN grown on GaN, the average total electron–longitudinal optical phonon scattering rate is \( (5.1 \pm 1.0) \times 10^{13} \text{ s}^{-1} \). This enormous electron–longitudinal optical phonon scattering rate, which is comparable to that observed in GaN, has been attributed to the extremely polar nature of InN. © 2007 American Institute of Physics. [DOI: 10.1063/1.2732174]

Recent development of crystal growth technique has produced high quality, single crystal InN that has a band gap of about 0.7 eV at room temperature.\(^1\)-\(^5\) This indicates that its ternary compound In\(_{1-x}\)Ga\(_x\)N not only has great potential for white light generation but also is suitable for applications in solar cells. On the device applications,\(^6\)-\(^10\) InN has been theoretically predicted and experimentally demonstrated to have enormously large transient electron drift velocity. As a result, InN is also very attractive for use in the fabrication of electronic devices of extremely high performance. Although considerable knowledge about the properties of InN has been obtained, dynamical information, such as that of the electron-phonon interaction, which is indispensable for designing high speed electronic devices, is still unknown. In this letter, we have used subpicosecond time-resolved Raman spectroscopy to interrogate electron–longitudinal optical (LO) phonon interactions in InN. We have found that, for a thick InN sample, the average total electron-LO phonon scattering rate is \( (5.1 \pm 1.0) \times 10^{13} \text{ s}^{-1} \).

The sample studied in this work has been described elsewhere (Ref. 11).

The experimental technique—time-resolved Raman spectroscopy—employed in this work has been described in detail somewhere else.\(^12\),\(^13\) The output of the second harmonic generation of a mode-locked Ti:sapphire laser was used as both the excitation and probing sources in our pump/probe experiments. The laser, which had a repetition rate of 80 MHz and a pulse width of about 100 fs, was chosen to operate at a wavelength of 442 nm. In the pump/probe configuration, the ultrashort pulse train was split into two beams of equal intensity but different polarizations. An appropriate analyzer was placed in front of the entrance of the spectrometer so that scattered light from the pump pulse was minimized while that from the probe pulse was allowed to be detected. The Raman signal was collected and analyzed by a standard computer-controlled Raman system which included a double spectrometer, a photomultiplier tube/charge coupled device, and its associated photon counting electronics. All the experimental data were taken at \( T=10 \text{ K} \). The photoexcited electron-hole pair density was estimated from the average laser power, the focused spot size on the sample surface, and the absorption depth at the excitation laser wavelength. The zero delay at the sample was determined to within \( \pm 0.01 \text{ ps} \) by the observance of the interference effect which occurred when the pump and probe pulses were spatially and temporally overlapped.

Figures 1(a) and 1(b) show typical Stokes and anti-Stokes Raman scattering spectra of the sample taken for two scattering configurations: \( X(Y,Z)Y \) and \( Z(X,X)Z \), as indicated, and at \( T=10 \text{ K} \) by the second harmonic of a cw mode-locked yttrium aluminum garnet (YAG) laser. The excitation laser has a pulse width of about 70 ps and a spectral width of \( \equiv 1 \text{ cm}^{-1} \). Here, \( X=(100) \), \( Y=(010) \), and \( Z \) is the direction parallel to the [0001] axis. The Raman modes are identified with the help of the Raman selection rules for wurzite semiconductors.\(^14\) The observation of LO phonon modes on the anti-Stokes side of Raman spectra at such a low temperatures as \( T=10 \text{ K} \) indicates that energetic electrons relax toward the bottom of conduction band primarily by emitting LO phonons.\(^15\) The most important aspect of these spectra is that under the \( X(Y,Z)Y \) scattering configuration, only the \( E_1(LO) \) phonon mode contributes to the anti-Stokes signal; whereas under the \( Z(X,X)Z \) geometry, only the \( A_1(LO) \) phonon mode. This intriguing aspect will be used in our time-resolved Raman experiments to measure the...
strength of electron-phonon interactions for the $E_1(LO)$ and the $A_1(LO)$ phonons, by the cw mode-locked Ti:sapphire laser, which inherently has a relatively broad spectral width (full width at half maximum $\approx 120$ cm$^{-1}$) because of the ultrashort pulse width (100 fs); in other words, we use $Z(X,Y)Z$ scattering configuration for the $A_1(LO)$ phonons and $X(Y,Z)Y$ for the $E_1(LO)$ phonons.

A typical integrated anti-Stokes Raman intensity, for the $A_1(LO)$ phonon mode as a function of time delay and with a photoexcited electron-hole pair density $n \approx 1 \times 10^{16}$ cm$^{-3}$, is shown in Fig. 2. The very rapid rise of the signal from around $\Delta t=0$ is a manifestation of an extremely large electron-LO phonon interaction in InN. It reaches a maximum at about 600 fs, indicative of the fact that at such a delayed time electrons are no longer emitting LO phonons that are detectable by our Raman spectroscopy. After about 600 fs, the anti-Stokes Raman intensity decreases with a decay constant of $\tau_{ph}=2.5\pm0.2$ ps.

To get better insight on electron-LO phonon interactions, we have used an electron cascade model$^{16,17}$ to fit the experimental data in Fig. 2.

In this model, the nonequilibrium LO phonon occupation number $n_{ph}(t)$ is given by the following rate equation:

$$\frac{dn_{ph}(t)}{dt} = G(t) - \frac{n_{ph}(t)}{\tau_{ph}}. \tag{1}$$

where $G(t)$ is the LO phonon generation rate by the excitation pulse laser and $\tau_{ph}$ is the LO phonon lifetime corresponding to the decay of the nonequilibrium phonons. For our current experimental conditions, the LO phonon generation rate is given by the approximation (the actual generation rate is a detailed function of the energy-dependent distribution function of the electrons at each step of the phonon cascade)

$$G(t) = \frac{f(t)}{\tau_{el-ph}}, \tag{2}$$

where $f(t) = 1$ for $0 \leq t \leq m\tau_{el-ph}$ and $f(t) = 0$ for $t > m\tau_{el-ph}$. $\tau_{el-ph}$ is the average electron-LO phonon scattering time, and $m$ is an integer determined by the excitation photon energy, LO phonon energy, band gap, and bandstructure of wurtzite InN.

For wurtzite InN, we use the energy-dependent effective mass $m_*(E)$ (Ref. 18) derived by a modified local-density approximation, which has been demonstrated to predict the correct band gap energy of InN (Refs. 1–5) and is also consistent with other measured electron effective masses around the $\Gamma$ valley. In addition, we take $m_0 = 1.63m_e$, $h\omega_L=2.81$ eV, $E_g=0.8$ eV, the index of refraction $n=2.9$, and $h\omega_{LO}=0.075$ eV [corresponding to the $A_1(LO)$ phonon energy]. However, due to conservation of both energy and momentum for the electron-LO phonon interaction process there exists a range of LO phonon wave vectors that electrons can emit. For an electron with wave vector $k_e$ and excess energy $\Delta E_e$, the minimum and maximum LO phonon wave vectors it can interact with are given by$^{21}$

$$k_{min} = \frac{\sqrt{2m_e}}{h} (\sqrt{\Delta E_e - \Delta E_e - h\omega_{LO}}), \tag{3}$$

and

$$k_{max} = \frac{\sqrt{2m_e}}{h} (\sqrt{\Delta E_e + \Delta E_e - h\omega_{LO}}). \tag{4}$$

Because of the nature of energy-wave vector relationship of the electron, the lower the electron’s energy, the larger the $k_{min}$ and the smaller the $k_{max}$. Therefore at some electron energy, the $k_{min}$ of LO phonon will be larger than the wave vector ($q=8.24 \times 10^5$ cm$^{-1}$) probed by our Raman scattering experiments. When that happens during the relaxation process, the energetic electrons can no longer emit LO phonons with wave vector detectable in our Raman scattering experiments. By taking this into consideration, we have found that although, in principle, the energetic electrons are capable of
emitting 25 LO phonons during their thermalization to the bottom of the conduction band, only 15 of them can be detected in our Raman experiments; in other words, $m = 15$ under our current experimental conditions.

Therefore, there are two adjustable fitting parameters in this electron cascade model: phonon lifetime ($\tau_{ph}$) and the average electron-phonon scattering time ($\tau_{el-ph}$). However, because the LO phonon lifetime $\tau_{ph}$ can be independently measured from the decaying part of the Raman signal, the average electron-LO phonon scattering time $\tau_{el-ph}$ is used as the only adjustable parameter in the fitting process.

We have found that $\tau_{el-ph} = 40 \pm 4$ fs best fit the experimental data in Fig. 2, which gives rise to an electron-LO phonon scattering rate of $\Gamma_{el-ph} = 1/\tau_{el-ph} = (2.5 \pm 0.3) \times 10^{13}$ s$^{-1}$.

Similar experiments for the $E_1$(LO) phonon mode were also carried out (which are not shown). Our experimental results in this case show that the average electron-LO phonon scattering rate for $E_1$(LO) phonon mode is given by $\Gamma_{el-ph} = 2.6 \pm 0.3 \times 10^{13}$ s$^{-1}$, which is very close to the value found for $A_1$(LO) phonon.

Therefore, the average total electron-LO phonon scattering rate in InN is given by $\Gamma_{total} = (5.1 \pm 1.0) \times 10^{13}$ s$^{-1}$. Since the average electron-LO phonon scattering rate in GaAs (Ref. 22) is about $5 \times 10^{12}$ s$^{-1}$, the observed average total electron-LO phonon scattering rate in wurtzite InN is almost one order of magnitude larger than that in GaAs and is comparable with what had been found for GaN.\textsuperscript{17}

The much larger TO-LO phonon energy splitting in wurtzite InN provides a clue to this mystery. We attribute this enormous increase of electron-LO phonon scattering rate in InN to its much larger ionicity. In general, the strength of electron-LO phonon coupling is set by the lattice-dipole interaction, which are expressed by\textsuperscript{18}

\[
1 = \frac{\omega^2_{LO}}{\gamma} \left[ \frac{1}{\varepsilon_c} - \frac{1}{\varepsilon_0} \right] = \frac{\omega^2_{LO} - \omega^2_{TO}}{\varepsilon_c},
\]

This splitting is directly proportional to the lattice polarization and is a measure of the effective charge. In GaAs, the LO-TO energy splitting is about 3 meV, while it is about 20 meV in wurtzite InN. This together with the much smaller dielectric constant of InN leads to an expected increase of a factor of about 10 in the electron-LO phonon scattering strength, which is quite close to that observed experimentally.

Indeed, in our previous Monte Carlo simulations of transport in InN,\textsuperscript{19} we found that the electron-LO phonon scattering rate is quite high, rising already to approximately $1.7 \times 10^{13}$ s$^{-1}$, for each of the two modes, at an energy of 150 meV (one $\hbar \omega_{LO}$ above the threshold for phonon emission), and assumes a value near $2.5 \times 10^{13}$ s$^{-1}$ over much of the higher energies in the nonparabolic central valley. This strong scattering process is a result of the large separation in $\varepsilon_0$ and $\varepsilon_c$ (12.0 and 7.3, respectively), as indicated above in Eq. (5).

In conclusion, we have used subpicosecond time-resolved Raman spectroscopy to study electron–longitudinal optical phonon interactions in InN. Our experimental results show that for a thick layer of InN grown on GaN, the average total electron–longitudinal optical phonon scattering rate is $(5.1 \pm 1.0) \times 10^{13}$ s$^{-1}$. We attribute this enormous electron-LO phonon scattering rate to the extremely polar nature of InN, and the measured value is in excellent agreement with values taken from Monte Carlo simulations using the actual calculated values for these scattering rates.

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\begin{thebibliography}{99}
\bibitem{20}Index of refraction for our sample is determined by an independent experiment.
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