Virginia Commonwealth University
VCU Scholars Compass

Electrical and Computer Engineering Publications | Dept. of Electrical and Computer Engineering

2006

Subpicosecond time-resolved Raman studies of LO phonons in GaN: Dependence on photoexcited carrier density

K. T. Tsen
Arizona State University at the Tempe Campus, tsen@asu.edu

Juliann G. Kiang
Walter Reed Army Institute of Research, Uniformed Services University of The Health Services

D. K. Ferry
Arizona State University at the Tempe Campus

Hadis Morkoç
Virginia Commonwealth University, hmorkoc@vcu.edu

Follow this and additional works at: http://scholarscompass.vcu.edu/egre_pubs

Part of the Electrical and Computer Engineering Commons

Tsen, K.T., Kiang, J.G., Ferry, D.K., et al. Subpicosecond time-resolved Raman studies of LO phonons in GaN: Dependence on photoexcited carrier density. Applied Physics Letters, 89, 112111 (2006). Copyright © 2006 AIP Publishing LLC.

Downloaded from
http://scholarscompass.vcu.edu/egre_pubs/113

This Article is brought to you for free and open access by the Dept. of Electrical and Computer Engineering at VCU Scholars Compass. It has been accepted for inclusion in Electrical and Computer Engineering Publications by an authorized administrator of VCU Scholars Compass. For more information, please contact libcompass@vcu.edu.

APPLIED PHYSICS LETTERS **89**, 112111 (2006)

Subpicosecond time-resolved Raman studies of LO phonons in GaN: Dependence on photoexcited carrier density

K. T. Tsen[a)]
Department of Physics and Astronomy, Arizona State University, Tempe, Arizona 85287

Juliann G. Kiang
Department of Cellular Injury, Walter Reed Army Institute of Research, Silver Spring, Maryland 20910-7500; Department of Medicine, Uniformed Services University of The Health Sciences, Bethesda, Maryland 20814-4799; and Department of Pharmacology, Uniformed Services University of The Health Sciences, Bethesda, Maryland 20814-4799

D. K. Ferry
Department of Electrical Engineering, Arizona State University, Tempe, Arizona 85287

H. Morkoç
Department of Physics, Virginia Commonwealth University, Richmond, Virginia 23284 and Electrical Engineering, Virginia Commonwealth University, Richmond, Virginia 23284

(Received 12 June 2006; accepted 18 July 2006; published online 12 September 2006)

Subpicosecond time-resolved Raman spectroscopy has been used to measure the lifetime of the LO phonon mode in GaN for photoexcited electron-hole pair density ranging from 10^{16} to 2×10^{19} cm^{-3}. The lifetime has been found to decrease from 2.5 ps, at low density, to 0.35 ps, at the highest density. The experimental findings should help resolve the recent controversy over the lifetime of LO phonon mode in GaN. © *2006 American Institute of Physics*.
[DOI: 10.1063/1.2349315]

A large nonequilibrium phonon population created in a semiconductor under the relaxation of carriers after excitation by an intense laser pulse, in particular, with the application of an electric field, not only causes a decrease of energy loss rate of the electronic system (the so-called "hot-phonon effects") but also reduces the electron drift velocity. These have been demonstrated and well understood in a variety of semiconductors such as GaAs and Si.[1] However, for some not so well established semiconductors, a case in point being GaN, the effects of the presence of a large nonequilibrium phonon population are less well understood, and sometimes the results are even contrary to our general belief.

The accumulation of a large nonequilibrium phonon population is a result of efficient electron-longitudinal optical phonon coupling and a relatively long phonon lifetime. Matulionis and co-workers[2–4] recently reported Monte Carlo simulations of hot-phonon effects in a biased GaN channel. To satisfactorily explain their experimental results, these authors found that the lifetime of longitudinal optical (LO) phonon which entered their complex model calculations as a fitting parameter needed to be about 350 fs. We note that this deduced value of LO phonon lifetime is about one order of magnitude shorter than that measured by time-resolved Raman spectroscopy.[5] Furthermore, it is significantly shorter than the value inferred from the frequency-domain measurements of Shi *et al.*[6] which set a lower bound on the LO phonon lifetime in GaN to be about 1 ps. On the other hand, recent calculations,[7,8] as well as experiments,[7] within our own group have confirmed the results of Matulionis *et al.*,[2] and illustrated the problem with understanding of the nonequilibrium phonon distribution in this wide band gap material. That is, the value of the phonon lifetime needed to match the experimental measurements in both clean heterojunction systems and in field-effect transistors is almost an order of magnitude lower than that found from the Raman scattering experiments. Matulionis [9] and Matulionis *et al.*[10] have subsequently suggested that there may be a carrier density dependence to the phonon lifetime. This raises a question: "what actually is the value of LO phonon lifetime in GaN?" To answer this important question, we have systematically measured the lifetime of "LO" phonon in GaN for a range of photoexcited electron-hole pair densities by subpicosecond time-resolved Raman spectroscopy. We find that the LO phonon lifetime is reduced with increasing carrier density, in a direct and systematic manner. If we refer a phononlike plasmon-LO phonon coupled mode to be the LO phonon, then all of the results in the literature are consistent with each other.

The GaN sample studied in this work was grown by metal organic chemical vapor deposition on a (0001)-oriented sapphire substrate. The GaN layer is about 6 μm thick and has a wurtzite structure. The sample is undoped and has a residual electron concentration of $<1 \times 10^{16}$ cm^{-3}. As depicted in Fig. 1, the third harmonic output of a cw mode-locked Ti-sapphire laser is used to excite and probe nonequilibrium phonons through Raman spectroscopy. The ultrafast laser source had a pulse width of about 80 fs and was chosen to operate at a repetition rate of 80 MHz and at photon energy of 5.0 eV. The nonequilibrium phonon populations were deduced in a manner similar to that of Ref. 11. The photoexcited electron-hole pair density was estimated from the average laser power, focused spot size on the sample surface, and the absorption depth at the excitation laser wavelength. All the data were taken at $T=300$ K. The double grating spectrometer used in this work has a spectral resolution of about 0.5 cm^{-1} and a spectral range from 22 000 to 62 000 cm^{-1}. One important advantage of probing

a)Electronic mail: tsen@asu.edu

This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to IP: 128.172.48.58 On: Mon, 13 Apr 2015 18:42:57

 © 2006 American Institute of Physics

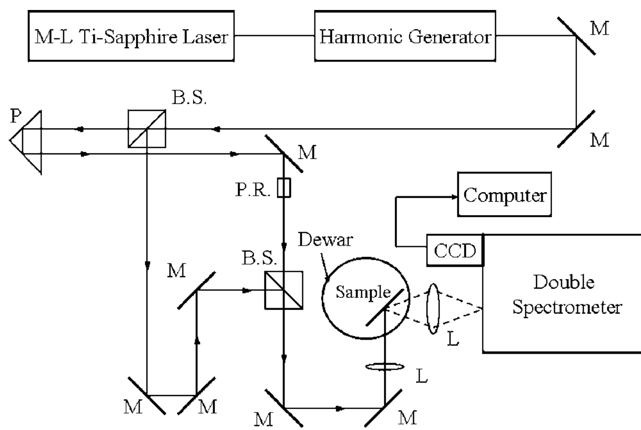

FIG. 1. Experimental setup used for subpicosecond time-resolved Raman measurements of LO phonons in GaN. M: mirror; BS: beam splitter; P: prism; PR: polarization rotator; L: lens; and CCD: charge coupled device.

nonequilibrium excitations with Raman spectroscopy in semiconductors is that since it detects a Raman signal only when excitation photons are present, the time resolution is essentially limited by the pulse width of the excitation laser and not by the response of the detection system. This explains why our detection system has a time resolution of the order of nanosecond, whereas the time resolution in our Raman experiments is typically on the scale of subpicosecond. Figure 2 demonstrates how the nonequilibrium optical phonons were generated. Electron-hole pairs are photoexcited by the excitation photons in the pump beam across the band gap of GaN, which has a band gap of about 3.42 eV. The energetic electron-hole pairs have an excess energy of about 1.58 eV and will rapidly relax to the bottom of the conduction band (for electrons) and to the top of the valence band (for holes) by emitting nonequilibrium phonons through electron-phonon interaction. For a polar semiconductor such as GaN, the strength of the Fröhlich interaction mechanism is much greater than that of any deformation potential mechanism.[12] As a result, these energetic carriers will relax by emitting LO phonons. By monitoring the occupation number of these emitted nonequilibrium phonons with the probe beam, information such as the strength of electron-phonon interactions and phonon-phonon interactions can be readily obtained.

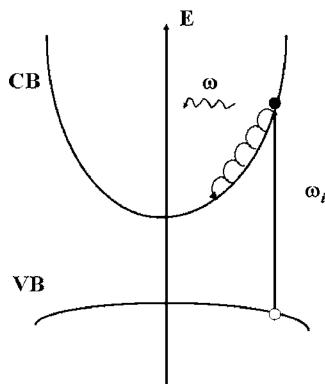

FIG. 2. Diagram demonstrating how the nonequilibrium LO phonons in GaN are created. Electron-hole pairs are generated by the excitation laser pulse in a direct band gap semiconductor. The nonequilibrium LO phonons are emitted (as indicated by the waved arrow near the letter ω) during the relaxation of energetic carriers. CB: conduction band; VB: valence band; E: energy; ω: LO phonon energy; and ω_i: excitation photon energy.

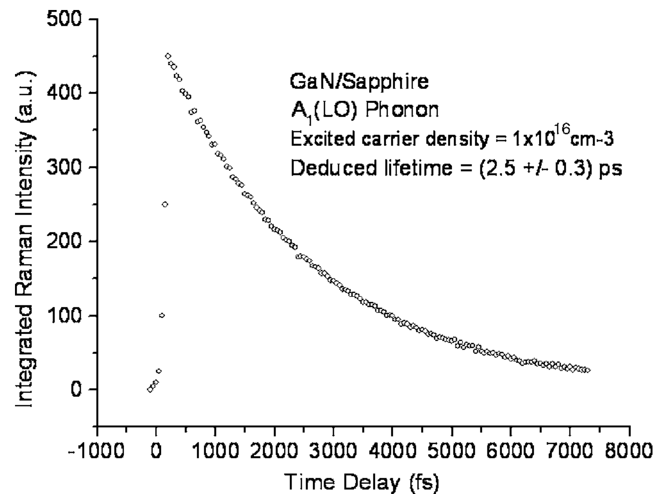

FIG. 3. Integrated Raman intensity of the A_1(LO) phonon as a function of the delay time for a GaN sample, with photoexcited electron-hole pair density of $n \cong 1x10^{16}$ cm^{-3}. The deduced lifetime of the LO phonon is found to be (2.5 ± 0.3) ps.

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 $n \cong 1 \times 10^{16}$ cm^{-3}, is shown in Fig. 3. The very rapid rise of the signal from around $\Delta t = 0$ is a manifestation of an extremely large electron-LO phonon interaction in GaN. It reaches a maximum at about 200 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 200 fs, the anti-Stokes Raman intensity decreases with a decay constant of $\tau = (2.5 \pm 0.3)$ ps. This we define as the population relaxation time or lifetime of the A_1(LO) phonon mode. The most likely decay channel for this phonon mode has been suggested by Tsen *et al.*[5] as a process in which a zone-center A_1(LO) phonon decays into a large wave vector TO and a large wave vector TA/LA phonon.

Similar experiments have been performed on the GaN sample with a range of photoexcited electron-hole pair densities by varying the spot size of the excitation laser. It is well-known[13] that electron plasma couples strongly with the LO phonon mode in polar semiconductors, in particular, for plasma density greater than 5×10^{17} cm^{-3}. GaN is no exception. We refer the phononlike plasmon-LO phonon coupled mode at high electron densities to be the LO phonon mode. Figure 4 shows the measured lifetime of this LO phonon mode in GaN as a function of the photoexcited electron-hole pair density, ranging from 10^{16} to 2×10^{19} cm^{-3}. We observe that the lifetime of this LO phonon mode decreases from 2.5 ps to about 0.35 ps, a result in keeping with the microwave noise measurements of GaN channels for heterostructure field-effect tranisistor,[9] since the electron density inside the channel typically is as high as 10^{19} cm^{-3}. We notice that a similar observation for GaAs has been reported by Kash and Tsang[14] and Tsang and Kash[15] for which the physical interpretation remained unclear.

In general, the lifetime is dominated by the decay of the LO phonons into a pair of acoustic phonons, as discussed above. This is generally a relatively slow process and cannot explain the density dependence. Moreover, screening cannot be invoked as a higher density would provide more screening and a slower response, opposite to that observed. However,

This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to IP: 128.172.48.58 On: Mon, 13 Apr 2015 18:42:57

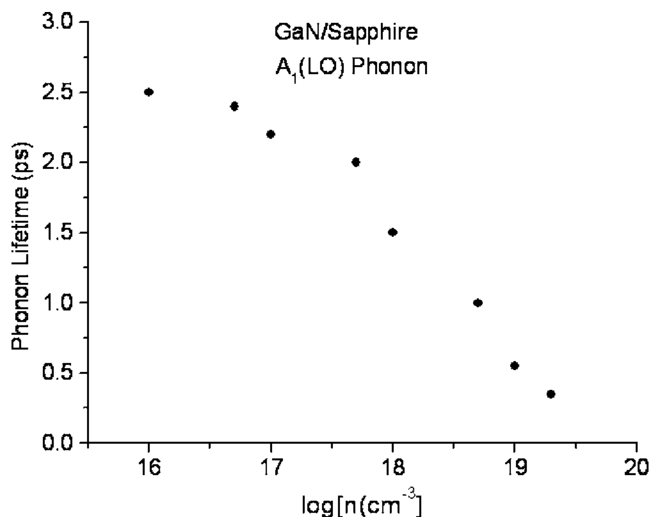

FIG. 4. A_1(LO) phonon lifetime as a function of the photoexcited electron-hole pair density as indicated. The lifetime has been found to decrease with increasing electron-hole pair density.

Matulionis has suggested that the hot LO phonons can emit a plasmon in the relaxation process.[9] This would explain the density dependence as well as a recent report of terahertz emission in a GaN field-effect device.[16] While intriguing, the mechanism for the decay via both plasmon and acoustic phonon modes has not yet been worked out, and more work is needed in this area.

In conclusion, subpicosecond time-resolved Raman spectroscopy has been used to measure the lifetime of LO phonon mode in GaN for photoexcited electron-hole pair density ranging from 10^{16} to 2×10^{19} cm^{-3}. The lifetime has been found to decrease from 2.5 to 0.35 ps. Our experimental findings help resolve the recent controversy over the lifetime of the LO phonon mode in GaN.

This work is supported by the National Science Foundation under Grant No. DMR-0305147. The work at VCU is supported by the Air Force Office of Scientific Research under the direction of K. Reinhardt. The authors would like to thank Steve Goodnick and Arvydas Matulionis for helpful discussions.

[1] For a review, see *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures*, edited by J. Shah, Vol. 115 in Solid State Sciences (Springer, New York, 1996), pp. 161–224.
[2] A. Matulionis, J. Liberis, I. Matulioniene, M. Ramonas, L. F. Eastman, J. R. Shealy, V. Tilak, and A. Vertiatchikh, Phys. Rev. B **68**, 035338 (2003).
[3] M. Ramonas and A. Matulionis, Semicond. Sci. Technol. **19**, S424 (2004).
[4] L. Ardaravicus, A. Matulionis, J. Liberis, O. Kiprianovicus, M. Ramonas, L. F. Eastman, J. R. Shealy, and A. Vertiatchikh, Appl. Phys. Lett. **83**, 4038 (2003).
[5] K. T. Tsen, D. K. Ferry, A. Botchkarev, A. Serdlov, A. Salvador, and H. Morkoc, Appl. Phys. Lett. **72**, 2132 (1998).
[6] L. Shi, F. A. Ponce, and J. Menendez, Appl. Phys. Lett. **84**, 3471 (2004).
[7] J. M. Barker, D. K. Ferry, S. M. Goodnick, D. D. Koleske, A. Allerman, and R. I. Shul, J. Vac. Sci. Technol. B **22**, 2045 (2004).
[8] S. Yamakawa, J. M. Barker, S. M. Goodnick, D. K. Ferry, and M. Saraniti, 27th International Conference on the Physics of Semiconductors (Flagstaff, AZ, 2005).
[9] A. Matulionis, Phys. Stat. Sol. **203**, 2313 (2006).
[10] A. Matulionis, L. F. Eastman, and J. Liberis, Proceedings of the International Conference on Hot Carriers in Semiconductors, 2005 (unpublished).
[11] E. D. Grann, K. T. Tsen, D. K. Ferry, A. Salvador, A. Botcharev, and H. Morkoc, Phys. Rev. B **56**, 9539 (1997).
[12] E. M. Conwell, *High Field Transport in Semiconductors* (Academic, New York, 1967).
[13] G. Abstreiter, M. Cardona, and A. Pinczuk, in *Light Scattering in Solids IV*, edited by M. Cardona and G. Güntherodt (Springer, New York, 1986), p. 5, and references therein.
[14] J. A. Kash and J. C. Tsang, Solid-State Electron. **31**, 419 (1988).
[15] J. C. Tsang, J. A. Kash, and S. S. Jha, Physica B & C **134**, 184 (1985).
[16] C. Skierbiszewski, K. Dybko, W. Knap, M. Siekacz, W. Krupczynski, G. Nowak, M. Bockowski, J. Lusakowski, Z. R. Wasilewski, D. Maude, T. Suski, and S. Porowski, Appl. Phys. Lett. **86**, 102106 (2005).

This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to IP: 128.172.48.58 On: Mon, 13 Apr 2015 18:42:57