1st International Symposium on Laser Ultrasonics: Science, Technology and Applications
July 16-18 2008, Montreal, Canada

Propagation of Multiple Coherent-Acoustic-Phonon Transients in GaN Single Crystals

Jie ZHANG,[1] Andrey BELOUSOV [2], Janusz KARPINSKI [2], and Roman SOBOLEWSKI [1]

[1]Department of Electrical and Computer Engineering and the Laboratory for Laser Energetics, University of Rochester, Rochester, NY, 14627-0231 USA; phone: +1 585 275 1808, fax: +1 585 273 1014;
e-mail: zhangj@ece.rochester.edu, roman.sobolewski@rochester.edu
[2]Solid State Physics Laboratory, ETH, CH-8093 Zurich, Switzerland
e-mail: belousov@phys.ethz.ch, karpinski@phys.ethz.ch

Abstract
We report our experimental investigations on generation, propagation, and detection of very long-lived, multiple, coherent-acoustic-phonon (CAP) picosecond transients in high-quality, ultra-thin (~53 μm) GaN single-crystal platelets. Our measurements were performed using a femtosecond two-color pump-probe spectroscopy technique, with 100-fs-wide, ultraviolet laser pump pulses used to generate the CAP bursts via inducing large electronic stress at the GaN surface. Subsequently, the phonon transients were detected and their propagation time-resolved by scanning the transient differential reflectivity signal of near-infrared (well below the GaN bandgap) probe pulses at both end surfaces of our sample. As a result, we observed sequential CAP transients, as well as their end-crystal reflections, each corresponding to a given pump-pulse excitation. The above experimental geometry allowed us not only to track the propagation of multiple phonon bursts inside our sample, but also yielded a direct measurement of the intrinsic CAP lifetime with negligible absorption and dispersion of the probe light. The amplitude of CAP oscillations was on the order of 10^{-5}–10^{-6}, and it was only dependent on the pump-beam absorption coefficient, while the CAP oscillation frequency was dispersionless, proportional to the probe light wave number, with the slope corresponding to the speed of sound in GaN.
Keywords: Coherent-acoustic-phonon, pump-probe spectroscopy, CAP lifetime

1. Introduction

Acoustic phonon oscillations in solids have most recently been investigated with a great interest, not only from the fundamental physics point of view, but also for their potential applications in photo-acoustic microscopy of nanostructures, THz acoustic-optic modulation, coherent sources for phonon spectroscopy, and, finally, nanoscale acoustic imaging [1-5]. In semiconductors, absorption of an ultrashort optical pulse generates picosecond in duration acoustic transients (coherent-phonon bursts) due to a variety of mechanisms, such as electric and/or thermal stress, deformation potential coupling, or the piezoelectric effect. Propagating phonons cause a periodic modulation of the optical properties of the semiconductor, which is usually detected by changes in reflectivity or transmissivity of delayed optical probe pulses. Coherent acoustic phonons (CAPs) in GaN films were first observed by Huang *et al.* [2] and, subsequently, were thoroughly investigated by Wu *et al.* [6, 7] in bulk GaN crystals. Using fixed (long) optical delays between the pump and probe beams, our group has demonstrated [8] that CAPs in GaN bulk samples can propagate macroscopic distances without losing their coherence in room temperature. In this work, we report on our observation of multiple coherent-acoustic-phonons transient in ultra-thin GaN single crystals and present new results on the intrinsic CAP lifetime with a modified experimental geometry.

2. Growth of GaN single crystals and experimental geometry

Our GaN bulk crystals were grown by a high-pressure solution-growth (HPSG) method [9]. The growth process was carried out at an external nitrogen gas pressure of 8-14 kbar and temperatures of 1350–1600 °C, because of high solubility of GaN in Ga at high temperatures. Nitrogen was first compressed with a commercial compressor up to 3 kbar and then compressed up to 10-15 kbar with a pressure intensifier. The pressure intensifier chamber had an internal diameter of 30 mm and in this chamber the high temperature furnace with a Pt-Rh heater was placed. Nitrogen was then transported into the metallic Ga melt with a temperature gradient of 5-50 °C/cm. GaN single crystals formed at the cooler zone of the HPSG chamber. They grew up as highly transparent platelets with sizes of up to 3×4 mm^2 and thicknesses varying from 50 μm to 1 mm.

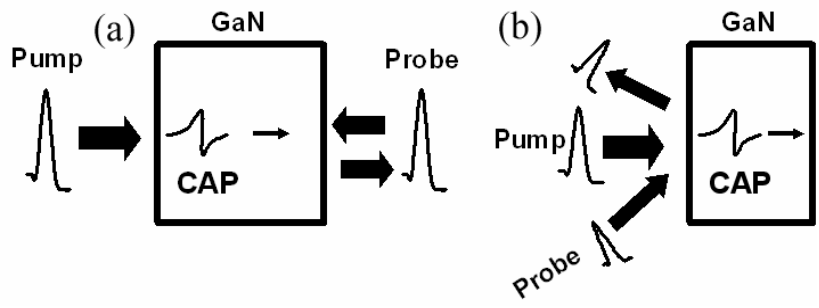

Figure 1. Schematic diagram of (a) two-sides and (b) one-side pump-probe CAP experiment.

The method for optical generation and time-resolved detection of CAPs is shown schematically in Fig. 1. Both one- and two-sides pump-probe spectroscopy experiments were performed on bulk GaN single crystals in the reflection mode. For excitation of coherent phonons, an all solid-state laser system was used. The system comprised a Verdi as the pumping laser and a commercial Ti:Sapphire mode locked laser, which produced a 76-MHz train of 100-fs-wide optical pulses at the wavelength range near 800 nm. With the aid of a home-made third-harmonic generator (THG), frequency tripled ultraviolet (UV) optical pulses were generated, with a pulse width of ~150 fs and photon energy of 4.64 eV (wavelength of 267 nm). These UV pulses were used as the pump focused onto the GaN surface with a spot diameter of ~20 μm and an incident influence ~0.04 mJ/cm^{-2} per pulse. Probe pulses, used to detect the CAP pulses, were the ones directly generated by the Ti:Sapphire laser. The probe pulses were at normal incident to the sample surface and had at least 10 times lower fluency than that of the pump. The excitation pulses were modulated at 2.5 kHz by a mechanical chopper. Time-resolved normalized reflectivity change $\Delta R/R$ of the probe beam was measured using a lock-in technique over a time scale of ~500 ps.

3. Generation and detection of CAPs

3.1 CAP pulse generation

The all-optical CAP generation and detection mechanisms in bulk GaN crystals were investigated in detail by Wu *et al*. [7], based on the seminal work of Thomsen *et al*. [1].

The approach to the CAP generation reduces to a one-dimensional (1-D) problem, based on the 1-D elastic wave equation:

$$\frac{\partial^2 \eta_{zz}(z,t)}{\partial t^2} - v_s^2 \frac{\partial^2 \eta_{zz}(z,t)}{\partial z^2} = \frac{\partial}{\partial z} S(z,t) \tag{1}$$

The driving force $S(z,t)$ is related to both electronic and thermal stresses [7]. Figure 2 presents an example of numerically computed profiles of CAP transients, generated by electronic stress, thermal stress, and the sum of both, respectively. The CAP wave packets were calculated in this case for the far-above-bandgap pump-photon excitation. We note that the CAP pulses have strength of the order of 10^{-5} and a spatial width of approximately twice the pump beam penetration depth.

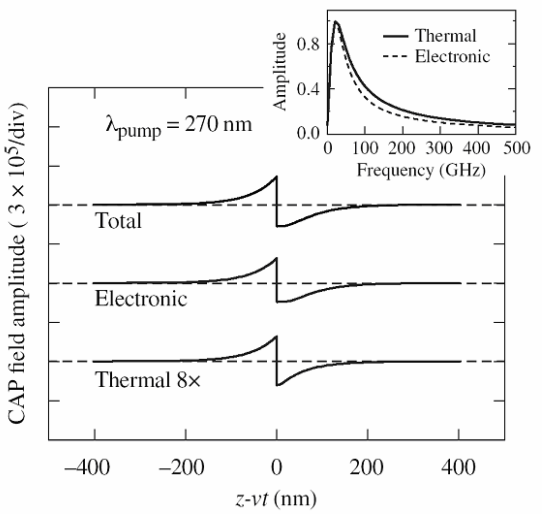

Figure 2. Simulation of spatial-time profiles of propagating CAP transients generated by the electronic stress, thermal stress, and both, respectively. The CAP pulse was excited by the pump beam with the photon energy of 4.59 eV. The inset shows the frequency spectra of the electronically (solid line) and thermal induced (dashed line) CAPs.

3.2 Detection of multiple CAPs

In our case, CAP pulses were generated in the near-surface sample region propagated into the sample along z-axis. An infrared probe pulse from the Ti:Sapphire laser was used to detect the CAP pulses. The probe beam, normally incident on the sample surface with a spot diameter ~10 μm (within the pump beam spot), generated two reflection beams: one corresponding to the partial reflection from the sample's surface and the second as the direct reflection from the propagating CAP burst. The interference between these two reflection beams produces the single-frequency oscillations on top of the experimentally observed probe's $\Delta R/R$ photoresponse and can be expressed as:

$$\frac{\Delta R}{R} = A \sin(2\pi f t - \varphi) e^{-t/\tau_d}, \tag{2}$$

where the amplitude A and the phase φ are the only pump-beam-energy dependent parameters and f is the frequency CAP oscillations. Since f comes from the self-inference of the probe light, reflected from the sample at near normal incidence, it is

given by

$$f = \frac{n\nu}{\pi} k_{probe} = \frac{2n\nu}{\lambda_{probe}} \ , \tag{3}$$

where ν is the speed of sound in GaN and $k_{probe} = 2\pi/\lambda_{probe}$ is the probe-light wave vector. Finally, τ_d is the experimental decay time and can be expressed as

$$\frac{1}{\tau_d} = \frac{1}{\tau_{phonon}} + \nu\alpha_{prb} \ , \tag{4}$$

where τ_{phonon} is the CAP intrinsic decay time, and α_{prb} is the probe light absorption coefficient.

Figure 3 shows the time-resolved differential $\Delta R/R$ from an ultra-thin (~55 μm) GaN crystal excited with the 267-nm pump pulses and probed with the 801-nm light, both illuminating the same side of our sample. Strong CAP oscillations are observed both before and after the zero delay. We note that the after-zero-delay CAP oscillations are imposed on top of the $\Delta R/R$ electronic (carrier relaxation) signal. To get a clear view of the oscillations, we subtracted the initial $\Delta R/R$ rise and the exponential decay from the raw data, as is shown in Fig. 3(b). The resulting oscillations exhibit the single frequency of 46.3 GHz, which is in full agreement with Eq. (3). Two insets of Fig. 3(a) show the same oscillating frequency obtained using the fast Fourier transform. The fact that we see CAPs both before and after the zero delay obviously indicates that we deal with multiple (at least two) propagating phonon bursts inside the sample at the same time: one generated by the current pump pulse (after the zero delay), while the other one from the previous pump pulse and detected before the zero-delay after being reflected back from the end side of the crystal.

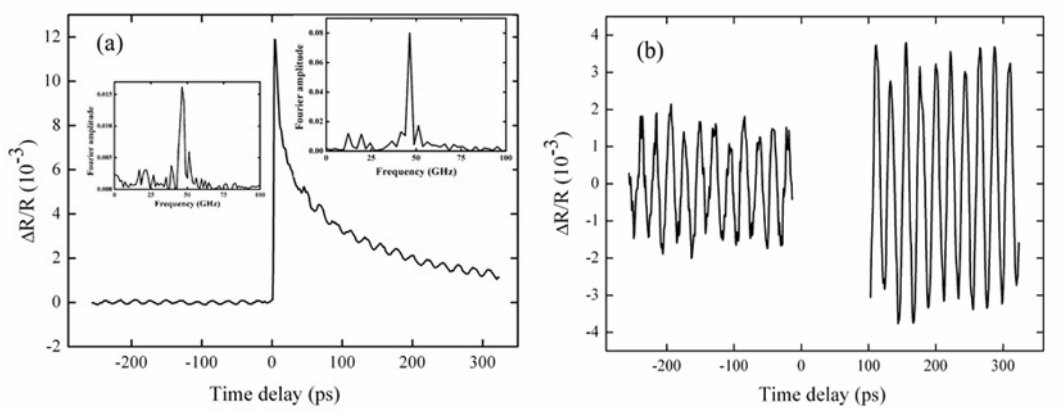

Figure 3. $\Delta R/R$ signal obtained using two-color pump-probe spectroscopy. (a) Full time-resolved $\Delta R/R$ transient. The insets in (a) are fast Fourier transforms (FFTs) of the oscillation before and after the zero-delay, respectively. Both have the same peak frequency at 46.3 GHz. (b) Two CAP oscillations corresponding to signals before and after the zero-delay point.

3.3 Intrinsic lifetime of CAPs

In a previous work [8], we have demonstrated that the ultra-long-lived CAPs have an intrinsic lifetime τ_{phonon} of at least 83 ns at room temperature, by investigating the CAP oscillations at longer time windows, using fixed optical delays between the pump and probe [10]. The actual τ_{phonon} value is likely to be longer, since our ~100 fs probe pulses not only attenuate, but also disperse as they propagate inside the GaN, which is another contribution to τ_d [11]. A modified experimental geometry, shown in Fig. 1(a), was adopted in order to eliminate the dispersion of the probe pulses. Two-sided pump-probe experiment was performed on a specially selected GaN platelet with a thickness of ~104 μm. We note that for v = 8002 m/s, it should take ~13 ns for a CAP to reach the end surface of our GaN sample. We also note that the 13-ns delay is for us practically the longest achievable value, because of the 76-MHz repetition rate (pulse separation of 13.2 ns) of our Ti:Sapphire laser. By exciting the CAP at one side and probing at the other side of our sample, two sets of oscillation data were obtained at 0 ns and 13 ns delay times, respectively, as shown in Fig. 4. The solid lines in Fig. 4, represent the fits based on Eq. (2) with the single decay constant τ_d = 88 ns. Since implementing our two-sided pump-probe spectroscopy, we eliminated propagation of the probe beam inside the crystal, we can simplify Eq. (4) and assume that τ_d = 88 ns is the actual τ_{phonon} of CAPs.

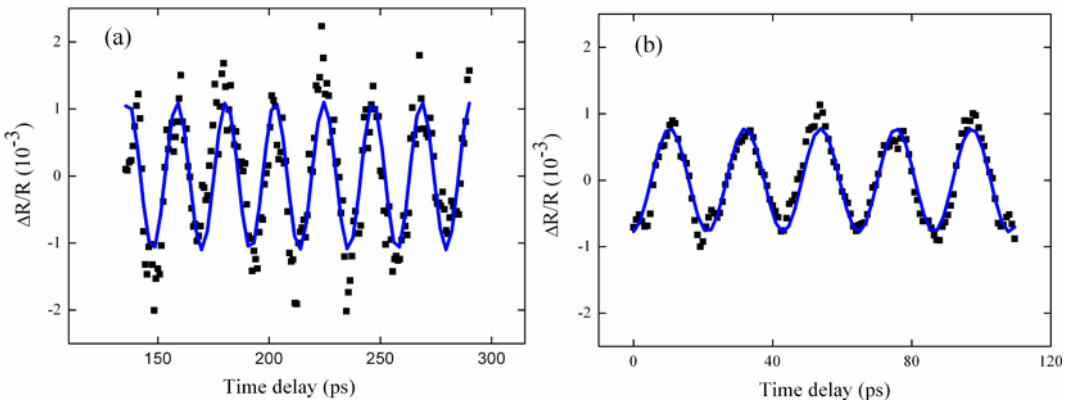

Figure 4. Δ*R*/*R* signal obtained using two-side pump-probe spectroscopy. (a) CAP oscillations (dots) within a 200-ps time window. (b) CAP oscillations measured after 13 ns time delay at the end surface of the crystal. The solid lines in (a) and (b) represent the fits based on Eq. (2).

4. Conclusions

We have studied CAP propagation in ultrathin bulk GaN platelets. The oscillations were initiated by the pump-beam induced electronic stress associated with the deformation potential. Both one-side and two-side pump-probe spectroscopy experiments were performed in two-color reflection mode. Propagation of multiple CAPs in GaN crystals was observed and analyzed. The intrinsic lifetime of GaN was determined to be ~88 ns under our modified experimental scheme, where CAPs were generated on the one side of the sample and detected on the other.

Acknowledgements

The authors would like to thank A. Cross for many valuable discussions throughout the course of this work. J. Z. acknowledges support from the Frank Horton Graduate Fellowship Program at the University of Rochester Laboratory for Laser Energetics, funded by DOE. The support of DOE does not constitute an endorsement by DOE of the views expressed in this article.

References

1. C. Thomsen, H. T. Grahn, H. J. Maris, and J. Tauc, *Phys. Rev. B* 34, pp 4129-382, 1986
2. Y. K. Huang, G. W. Chen, and C. K. Sun, *Appl. Phys. Lett.* 79, pp 3361- 63, 2001
3. O. Matsuda, O. B. Wright, D. H. Hurley, V. E. Cusev, and K. Shimizu, *Phys. Rev. Lett.* 93, pp 095501, 2004
4. C. K. Sun, J. C. Liang, and X. Y. Yu, *Phys. Rev. Lett.* 84, pp 179, 2000
5. B. C. Daly, N. C. R. Holme, T. Buma, et al. Appl. Phys. Lett. 84, pp 5180-5182, 2004
6. S. Wu, P. Geiser, J. Jun, J. Karpinski, J. R. Park and R. Sobolewski, *Appl. Phys. Lett.* 88, pp 041917, 2006
7. S. Wu, P. Geiser, J. Jun, J. Karpinski, and R. Sobolewski, *Phys. Rev. B* 76, pp 085210, 2007
8. S. Wu, J. Zhang, A. Belousov, J. Karpinski, and R. Sobolewski in Gallium Nitride Materials and Devices III, ed. by H. Morkoç, C. W. Litton, J.-I. Chyi, Y. Nanishi, and E. Yoon, *Proc. of SPIE*, 6894, 68940K, 2008
9. J. Karpinski, J. Jun, and S. Porowski, *Jour. Cryst. Growth* 66, pp 1-10, 1984
10. S. Wu, J. Zhang, A. Belousov, J. karpinski, and R. Sobolewski, *Journal of Physics Conference Series* 92, pp012021, 2007
11. H. N. Lin, R. J. Stoner, H. J. Maris, and J. Tauc, *J. Appl. Phys.* 69(7), pp 3816-3822, 1991