

Narrow-band detection of propagating coherent acoustic phonons in piezoelectric InGaN/GaN multiple-quantum wells

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The authors demonstrated that the piezoelectric superlattice, can serve as narrow-band detectors for propagating coherent longitudinal acoustic phonons at multiple frequencies corresponding to the spatial frequency of the superlattice and its higher harmonics, and its detection bandwidth is determined by the total structure width. By optically exciting a broadband propagating longitudinal acoustic pulse from a thin Ni film, the authors studied the acoustic spectral sensitivity function of a ten-period In_{0.12}Ga_{0.88}N/GaN multiple quantum well. Because the barriers (19 nm) and wells (3.6 nm) are of different widths, the second detection band, corresponding to the second harmonic of the fundamental frequency, can be resolved. © 2007 American Institute of Physics.

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It has been shown that ultrafast optical excitation of GaAs/AlAs and GaAs/Al_xGa_{1-x}As superlattice structures^{1,2} and piezoelectric InGaN/GaN multiple quantum wells^{3,4} (MQWs) results in the generation of coherent acoustic phonons (CAPs). However, in these previous experiments the semiconductor structures were not only a generator but also a detector of the CAPs, and few experiments were conducted where the generation and the detection of CAPs were spatially separated.^{5,6} For piezoelectric superlattices, its spectral sensitivity function for coherent acoustic phonon detection has never been independently measured.⁷ This motivated us to generate broadband acoustic phonons in a separated layer^{8,9} to investigate the detection sensitivity of a periodic piezoelectric multilayer. In this paper, we demonstrate that the piezoelectric superlattice with a low impedance mismatch between adjacent layers can serve as narrow-band CAP detectors at multiple frequencies corresponding to the spatial frequency of the MQWs and its higher harmonics. We adopt In_{0.12}Ga_{0.88}N/GaN MQW as our studied narrow-band CAP detectors. With a broad-spectrum propagating longitudinal acoustic pulse (LAP), which resulted from an ~3–6 nm Ni film after optical impulse excitation, we studied the acoustic spectral sensitivity function for optical transmission of the ten-period In_{0.12}Ga_{0.88}N/GaN MQW. With an asymmetric well/barrier ratio and a broad enough LAP bandwidth, the InGaN/GaN MQW experimentally exhibited two narrow detection bands of which the central frequencies correspond to the period of the MQW and its second harmonic, respectively,¹⁰ and the CAP detection bandwidth is determined by the total MQW width.

In periodic semiconductor multilayer structures, due to the zone folding of the phonon branches into mini-Brillouin zones with a dimension of π/Λ and the impedance mismatch between layers, the phonon stop bands appear at acoustic wave vectors of $q=n\pi/\Lambda$, where Λ is one-period width of the multilayer structure and n is an integer. The acoustic phonons with a stop band frequency cannot propagate in these structures and will be reflected due to the Bragg condition.¹¹ In previous works, phonon filters based on stop bands in periodic semiconductor multilayers have been realized.^{12–15} As a result, when the propagating LAP incidents upon the periodic semiconductor multilayers with a large acoustic impedance mismatch, it will be hard for this structure to detect propagating phonons with a stop band frequency. In contrast to the previous stop band filter result,^{12–15} with a low impedance mismatch between piezoelectric multilayers with a limited number of layers, for example, the ten-period In_{0.12}Ga_{0.88}N/GaN MQW,^{16,17} we expect that the piezoelectric multilayer structure will not only allow transmission of the acoustic phonons with an acoustic wave vector of $q=2n\pi/\Lambda$ with negligible interfacial reflections but can also provide narrow-band CAP detection through the strain induced modulation in the quantum confined potentials,^{4,7,18} with the central frequency of detection corresponding to $q=2n\pi/\Lambda$ and its higher harmonics.

The sample we used in this experiment was grown on an ~3.4- μ m-thick wurtzite GaN layer on a *c*-axis sapphire substrate by metal organic chemical vapor deposition. It has ten periods of *c*-axis orientated In_{0.12}Ga_{0.88}N/GaN (3.6/19 nm) layers as the narrow-band CAP detector, and the structure was confirmed by transmission electron microscopy, as shown in Fig. 1. The room temperature photoluminescence of the MQW is with a peak at ~460 nm. On top of the MQW, the sample was capped with a 17 nm layer of unintentionally doped GaN. Finally, we deposited an ~3–6 nm

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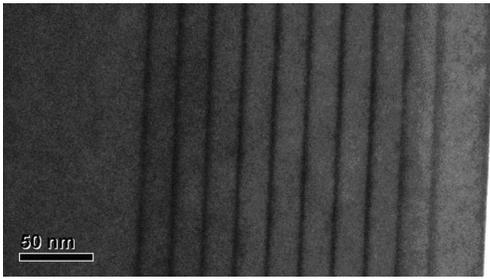


FIG. 1. Transmission electron micrograph of the ten-period $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}/\text{GaN}$ MQW. The first layer on right hand side includes the ~ 17 -nm-thick undoped GaN cap layer of and one ~ 19 -nm-thick barrier layer. The left hand side shows one part of the $\sim 3.4 \mu\text{m}$ GaN layer. The quantum well width is ~ 3.6 nm and the barrier width is ~ 19 nm.

Ni film on top of the cap layer as a generator of LAP, and the uncertainty of the Ni film thickness came from limited deposition thickness control of the thermal coater. Within the thickness uncertainty, the Ni film can generate broad-spectrum acoustic phonons at least up to 800 GHz, enough for our current study. Because the acoustic impedance of Ni is similar to that of GaN ($Z_{\text{Ni}}/Z_{\text{GaN}} \sim 1.09$),^{16,17,19} the acoustic reflection coefficient between the Ni film and the undoped GaN cap layer is less than 0.05.

The experimental setup is a femtosecond transient transmission two-color pump-probe system.²⁰ A femtosecond Ti:sapphire laser at a 76 MHz repetition rate provided optical pulses at a central wavelength of 800 nm for optical pumps. Pulses frequency doubled by a Beta-Barium Borate crystal, now at a central wavelength of 400 nm, were used as optical probes. The powers of the pump and the probe beams were adjusted to be 40 and 6.5 mW, respectively. According to previous experimental^{4,18} and theoretical⁷ works on coherent LA phonon oscillations in piezoelectric MQWs, the strain induced modulation in quantum confined energies can be through several mechanisms, including piezoelectric coupling and deformational potential coupling. A detailed theoretical investigation⁷ found that the piezoelectric coupling is the dominating contribution for optical transmission sensitivity function in $\text{In}_{0.1}\text{GaN}$ MQWs. By choosing the probe photon energy above the InGaN quantum well transition but below the bandgap of the GaN (~ 365 nm), similar to previous studies,⁴ we are able to probe the strain induced absorption changes in the quantum well regions. The temporal delays between the pump and the probe pulses were controlled by a translation stage. Finally, the pump and the probe beams were focused onto the sample with spot diameters of ~ 20 and $\sim 10 \mu\text{m}$, respectively, and the transmitted probe pulses were detected by a photodetector linked to a lock-in amplifier. After the Ni film was excited by the pump radiation, a propagating LAP was generated and went through the 17 nm undoped GaN cap layer into the MQW region along the c axis. The absolute amplitude of the strain pulse can be estimated to be on the order of 3×10^{-4} , following Ref. 21. The propagating LAP in the quantum well modulated the piezoelectric field and deformation potential of the InGaN well and caused variation on optical absorption of the probe beam.^{4,7} The transmission changes of the probe beam due to the existence of coherence LA phonons can be conveniently expressed using this equation $\Delta T(t) = \mathcal{F}^{-1}[F(\omega)S(\omega)]$, where \mathcal{F}^{-1} denotes the inverse Fourier transform and $F(\omega)$ and $S(\omega)$ are the acoustic spectral sensitivity function for optical transmission of the MQW and the frequency spectrum of the

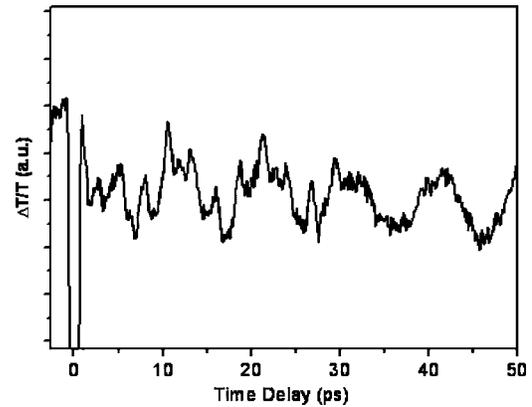


FIG. 2. Measured transient transmission change vs probe time delay.

strain pulse, respectively. The quantity $F(\omega)$ represents the weighted contribution of the strain value to the sample transmission change at a specific acoustic angular frequency ω , and it is related to the wavelength of the probe and the structure of the sample. As shown in Fig. 2, we can see the transmission variation of the probe beam when the LAP propagates into the sample. According to the velocities of the LA modes of Ni and GaN along the c axis,^{16,17,19} we can know that the transient transmission changes for the time delay of ~ 4 –30 ps is resulted from the LAP propagating in the $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}/\text{GaN}$ MQW region. Hence we use the transient transmission signal between 4 and 30 ps to study the characteristic of the acoustic phonon spectral sensitivity function $F(\omega)$ of the ten-period $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}/\text{GaN}$ MQW.

In order to investigate its frequency domain response, we performed fast Fourier transformation on the transient transmission signal shown in Fig. 2 for ~ 4 –30 ps, and the result is shown in Fig. 3(b). There are three signal bands in Fig. 3(b) with central frequencies of ~ 103 , ~ 370 , and ~ 740 GHz and with full widths at half maximum of ~ 50 , ~ 45 , and ~ 45 GHz, respectively. The ~ 370 GHz frequency corresponds to the acoustic wave vector $q = 2\pi/\Lambda$, where $\Lambda = 22.6$ nm, assuming the longitudinal acoustic sound velocity along the c -axis $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}/\text{GaN}$ MQWs to be 8360 m/s, slightly higher than the reported value of 8020–8160 m/s for bulk GaN (Refs. 22 and 23) and is in good agreement with a recently reported value in InGaN/GaN MQW.²⁴ Furthermore, since the widths of the

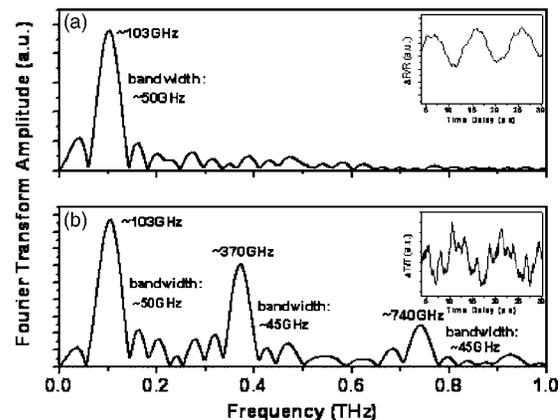


FIG. 3. (a) Fourier transform of the inset of (a), which shows the measured transient reflection change for ~ 4 –30 ps probe time delay. The absolute magnitude of the relative reflectivity oscillation $\Delta R/R$ is on the order of 3×10^{-5} . (b) Fourier transform of the inset of (b), which shows the measured transient transmission change for ~ 4 –30 ps probe time delay.

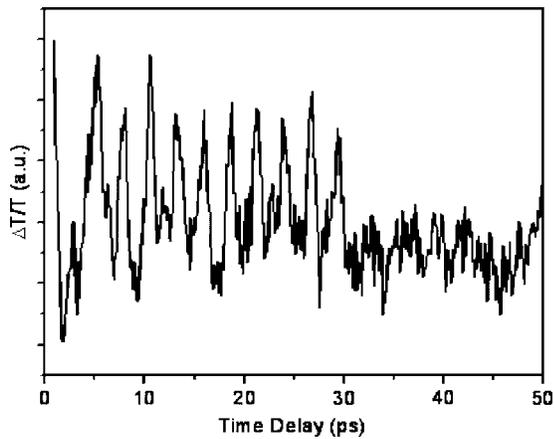


FIG. 4. Measured transient transmission trace with the band at a central frequency of ~ 103 GHz filtered out. Ten significant peaks, situated between ~ 4 and 30 ps, correspond to the time when the LAP propagating through the ten equally spaced InGaN quantum wells.

barriers and that of the wells are not the same ($19/3.6$ nm), in principle, the acoustic spectral sensitivity function of the MQWs has multiple detection bands. With an asymmetric well/barrier ratio, the MQW spatial frequencies are with a fundamental wave vectors of $q=2\pi/\Lambda$ and its higher harmonics.¹⁰ With a limited bandwidth phonon source $S(\omega)$, we were able to observe the fundamental frequency of ~ 370 GHz and its second harmonic, i.e., ~ 740 GHz, being detected by the $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}/\text{GaN}$ MQW. Moreover, in theory, the bandwidth of these multiple detection bands must be the same, which are determined by the total MQW width D . The bandwidths of ~ 370 GHz band and ~ 740 GHz band are ~ 45 GHz, which also consists with the expectation that the bandwidth should be equal to $\sim 1.2v_a/D$, where v_a is the acoustic velocity in the MQW.

As regards the ~ 103 GHz signal, we must first understand the detection mechanism of picosecond ultrasonics in transparent (or partially transparent) materials.^{21,25,26} Its detection mechanism is based on the modification of the optical properties in strained materials due to a LAP inside. In previous picosecond ultrasonics studies, the transient reflectivity of the probe beam oscillates when the LAP propagates in the medium, which is the so-called “Brillouin oscillation.”²⁶ When the optical reflection varies in a transparent system, the optical transmission must change at the same time; therefore, the intensity of the transmitted probe beam also vacillates corresponding to the Brillouin oscillation, and both oscillations have the same frequency. For this reason, we suggest that the ~ 103 GHz transmission signal comes from the Brillouin oscillation. To verify this, we performed the corresponding transient-reflection two-color pump-probe experiment on the same sample, and the results are shown in Fig. 3(a). There indeed is an ~ 103 GHz oscillation with an ~ 50 GHz bandwidth, while the observed Brillouin oscillation is with a 180° phase shift from that observed in the transmission measurement, supporting our suggestion.

In order to exclusively study the characteristic of the $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}/\text{GaN}$ MQW acoustic detector in the time domain, we filtered out the ~ 103 GHz acoustic signals and the result is shown in Fig. 4. There are obviously ten peaks with a period of ~ 2.67 ps, corresponding to the LAP propagating through the ten equally spaced InGaN quantum wells one by

one. If there was strong acoustic reflection between $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}$ and GaN interfaces, the trace would have more than ten significant peaks. This result suggests that the propagating LAP is insignificantly affected by $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}/\text{GaN}$ multilayers, and the original profile of the LAP is basically kept during its propagation through the InGaN MQW.

In summary, we demonstrated that the low-impedance-mismatch piezoelectric superlattice can serve as narrow-band detectors for propagating longitudinal CAPs at multiple frequencies corresponding to the fundamental spatial frequency of the superlattice and its higher harmonics, and its detection bandwidth is determined by the total structure width. By optically exciting a broadband propagating longitudinal acoustic pulse from a thin Ni film, we studied the acoustic spectral sensitivity function for optical transmission of a ten-period $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}/\text{GaN}$ MQW. Because the barriers and wells are of different width, the second detection band, corresponding to the second harmonic of the fundamental frequency, can be resolved.

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- ¹A. Yamamoto, T. Mishina, Y. Masumoto, and M. Nakayama, *Phys. Rev. Lett.* **73**, 740 (1994).
- ²A. Bartels, T. Dekorsy, H. Kurz, and K. Köhler, *Phys. Rev. Lett.* **82**, 1044 (1999).
- ³C.-K. Sun, J.-C. Liang, C. J. Stanton, A. Abare, L. Coldren, and S. P. DenBaars, *Appl. Phys. Lett.* **75**, 1249 (1999).
- ⁴C.-K. Sun, J.-C. Liang, and X.-Y. Yu, *Phys. Rev. Lett.* **84**, 179 (2000).
- ⁵O. Matsuda, T. Tachizaki, T. Fukui, J. J. Baumberg, and O. B. Wright, *Phys. Rev. B* **71**, 115330 (2005).
- ⁶K. Mizoguchi, M. Hase, S. Nakashima, and M. Nakayama, *Phys. Rev. B* **60**, 8262 (1999).
- ⁷G.-W. Chern, K.-H. Lin, and C.-K. Sun, *J. Appl. Phys.* **95**, 1114 (2004).
- ⁸C. Thomsen, J. Strait, Z. Vardeny, H. J. Maris, J. Tauc, and J. J. Hauser, *Phys. Rev. Lett.* **53**, 989 (1984).
- ⁹C. Thomsen, H. T. Grahn, H. J. Maris, and J. Tauc, *Phys. Rev. B* **34**, 4129 (1986).
- ¹⁰G.-W. Chern, K.-H. Lin, Y.-K. Hung, and C.-K. Sun, *Phys. Rev. B* **67**, 121303 (2003).
- ¹¹S. Tamura, D. C. Hurley, and J. P. Wolfe, *Phys. Rev. B* **38**, 1427 (1988).
- ¹²V. Narayanamurti, H. L. Störmer, M. A. Chin, A. C. Gossard, and W. Wiegmann, *Phys. Rev. Lett.* **43**, 2012 (1979).
- ¹³N. M. Stanton, R. N. Kini, A. J. Kent, M. Henini, and D. Lehmann, *Phys. Rev. B* **68**, 113302 (2003).
- ¹⁴N. D. Lanzillotti-Kimura, A. Fainstein, A. Lemaître, and B. Jusserand, *Appl. Phys. Lett.* **88**, 083113 (2006).
- ¹⁵S. Mizuno and S. Tamura, *Phys. Rev. B* **45**, 13423 (1992).
- ¹⁶A. F. Wright, *J. Appl. Phys.* **82**, 2833 (1997).
- ¹⁷V. W. L. Chin, T. L. Tansley, and T. Osatchan, *J. Appl. Phys.* **75**, 7365 (1994).
- ¹⁸K.-H. Lin, G.-W. Chern, Y.-K. Huang, and C.-K. Sun, *Phys. Rev. B* **70**, 073307 (2004).
- ¹⁹B. A. Auld, *Acoustic Fields and Waves in Solids* (Wiley, New York, 1973), Vol. 1, p. 358ff.
- ²⁰C.-K. Sun, F. Vallée, L. H. Acioli, E. P. Ippen, and J. G. Fujimoto, *Phys. Rev. B* **50**, 15337 (1994).
- ²¹H.-N. Lin, R. J. Stoner, H. J. Maris, and J. Tauc, *J. Appl. Phys.* **69**, 3816 (1991).
- ²²C. Deger, E. Born, H. Angerer, O. Ambacher, M. Stutzmann, J. Hornsteiner, E. Riha, and G. Fischerauer, *Appl. Phys. Lett.* **72**, 2400 (1998).
- ²³Y.-K. Huang, G.-W. Chern, C.-K. Sun, Y. Smorchkova, S. Keller, U. Mishra, and S. P. DenBaars, *Appl. Phys. Lett.* **79**, 3361 (2001).
- ²⁴Ü. Özgür, C.-W. Lee, and O. Everitt, *Phys. Status Solidi B* **228**, 85 (2001).
- ²⁵O. B. Wright, *J. Appl. Phys.* **71**, 1617 (1992).
- ²⁶A. Devos and R. Côte, *Phys. Rev. B* **70**, 125208 (2004).