Generation of coherent acoustic phonons in strained GaN thin films

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Coherent acoustic phonon oscillations were generated and studied in strained GaN thin films. Inside the bulk GaN film, the longitudinal interference of an ultraviolet femtosecond pump pulse created periodic carrier distribution that screened out the strain-induced piezoelectric field and initiated the coherent longitudinal acoustic phonon oscillations corresponding to the carrier periods. The created coherent phonon oscillation modulated the piezoelectric field thus modified the absorption property of the GaN thin film through Franz–Keldysh effect. This time-dependent absorption modulation was reflected in the transmission variation of the followed probe pulses, resulting a long decay time ~ 300 ps for the initiated coherent phonon oscillations. © 2001 American Institute of Physics. [DOI: 10.1063/1.1418450]

Progress in femtosecond lasers and ultrafast spectroscopy technology has enabled us to generate and directly observe the coherent oscillation of phonon modes. Optically excited coherent phonon mode can behave like a classical oscillator and have nonzero time-dependent displacement. The corresponding modulation of the material dielectric constant could then be observed by changes in the intensities of transmitted or reflected probe light pulses. In semiconductors, carriers were excited by ultrashort pump pulses in a spatial area much larger than one lattice unit cell and the excited carrier populations (with wave vector $q \approx 0$) are coupled to the corresponding optical phonon modes (also with $q \approx 0$).^{1,2} For acoustic phonon excitations, due to wave vector constraint, only low frequency (low q) acoustic wave was directly excited in bulk materials. Higher frequency coherent acoustic phonon oscillations were previously observed in artificial semiconductor heterostructures such as AlAs/GaAs superlattices,^{3,4} GaAs/Al_xGa_{1-x}As and Ge/Si multiplayer structures,^{5,6} and PbTe and PbS quantum dots.^{7,8} Due to the artificial structures, acoustic waves corresponding to the structure periodicity or zone-folded acoustic phonon modes^{3,4} were observed. Recently we demonstrated coherent acoustic phonon oscillation in InGaN/GaN multi-quantum wells (MQWs), with phonon oscillation frequency tuned by MQW period width.⁹ Different from most previous studies, piezoelectric field inside the MQW was used to initiate the observed displacive oscillation. Due to the modulation of piezoelectric field properties, strong transmission variation on the order of 1% can be generated and observed. Even though the specific phonon mode was suggested to be selected by the coupling between the periodic carrier distribution and the corresponding acoustic phonon mode, there are discussions about the unavoidable contribution of zone-folded acoustic mode in InGaN MQWs.¹⁰

One way to generate $q \neq 0$ coherent longitudinal acoustic phonon oscillation without any contributions of zone-folded mode from artificial periodic structures is to generate coherent phonon oscillations directly in bulk materials. In this letter, we report the generation of coherent longitudinal acoustic phonon oscillations in strained GaN thin films with longitudinal interferometric techniques. The principle of the reported coherent acoustic phonon oscillation is similar to previous traveling coherent acoustic wave generation using laser induced gratings.^{11,12} Surface acoustic wave was previously generated by lateral interference of two excitation laser pulses with stimulated Brillouin scattering mechanisms. Thus demonstrated acoustic wave had frequency from 30 MHz up to 2 GHz, with a possible acoustic frequency up to 30 GHz proposed with a counter-propagating excitation scheme. In our experiment of coherent acoustic phonon excitation, it is the longitudinal self-interference of an ultraviolet femtosecond pump pulse that created periodic carrier distribution in a strained GaN thin film. This periodic carrier distribution screened out the local piezoelectric field, and a displacive coherent longitudinal acoustic phonon oscillation was initiated corresponding to the photoexcited carrier period width, with a momentum q in the sample growth direction perpendicular to the surface. Combining UV excitation, the method of longitudinal interference, and the nature of high sound velocity in GaN c axis, coherent acoustic phonons induced in the demonstrated experiment can have frequency higher than 100 GHz, which can be tuned by the pumping wavelength. With a much thicker total layer thickness compared with previous MQW samples, oscillation decay time longer than 200 ps can be observed.

The experiments were performed on a 0.60- μm -thick strained GaN thin film grown on top of an AlGaIn substrate. After annealing the c -plane sapphire substrate at 1050 °C, a 20-nm-thick buffer layer of GaN was deposited using metal-organic chemical vapor deposition (MOCVD). The temperature of the MOCVD was then raised to grow a 1.8- μm -thick

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FIG. 1. Transmitted probe signal with a pump/probe wavelength of (a) 390 nm and (b) 370 nm. Also noted is that a 180° phase difference existed in the oscillations of the two traces, as shown in the inset of (a) by differentiating the two traces.

AlGaIn layer. The Al content was around 5%. The unintentionally doped GaN layer with a thickness of $0.60\ \mu\text{m}$ was then grown on top of the AlGaIn substrate layer using molecular beam epitaxy. Room temperature photoluminescence measurements indicate a 3.42 eV (362 nm) band gap energy for the GaN layer and a 3.53 eV band gap energy for the AlGaIn substrate. A conventional transmission-type pump-probe setup was used in the room-temperature experiment. Femtosecond pulses with a pulse width around 150 fs were generated using a Ti sapphire laser. The output pulse was frequency doubled by a BBO crystal of 0.5-mm-thick to the UV wavelength range. The UV beam, with a pulse width of 250 fs, was then separated into a pump and a probe that would both be focused on the sample with a spot size around $7\ \mu\text{m}$. With a normal incident pump beam, self-longitudinal interference can be generated inside the GaN sample due to high interface reflection. With photoexcitation around the band tail states,¹³ periodic carrier distribution corresponding to the optical interference pattern can be generated with the resulted transmission change detected by the probe beam at the same wavelength. A series of probe transmission measurements with different pump/probe wavelengths, from 370 to 390 nm, were recorded.

When we photoexcited the bulk sample with 390 nm wavelength, coherent acoustic phonon oscillation induced transmission modulation was observed as shown in Fig. 1(a). With a pump fluence of $1.9 \times 10^{-5}\ \text{J}/\text{cm}^2$ and an average photocarrier density of $8.4 \times 10^{17}\ \text{cm}^{-3}$ in GaN, transmission modulation larger than 0.01% can be observed. It is interesting to notice its cosinusoidal nature, which indicates a dispersive oscillation. With 390 nm wavelength and 2.6 index of GaN,¹⁴ the pump self-longitudinal interference spacing was 75 nm, which was half of the UV pump wavelength inside sample. For this particular interference pattern, an oscillation frequency of 106 GHz was observed. Figure 1(b) demonstrates the initiated coherent acoustic phonon oscillations by pumping at a wavelength of 370 nm. With a shorter

FIG. 2. Dispersion curve of coherent longitudinal acoustic phonon shows a c -axis sound velocity of GaN about $8160 \pm 200\ \text{m/s}$.

interference pattern spacing of 71 nm, a higher oscillation frequency of 117 GHz could be observed. By performing the experiment with different pump wavelengths between 370 and 390 nm, a longitudinal acoustic phonon dispersion curve in bulk GaN c axis can be obtained. In Fig. 2 we find a linear relation between the phonon wave vector (which is the inversion of interference pattern spacing times 2π by Fourier series analysis) and the observed phonon oscillation frequency. Estimating the slope by a linear fitting will give a bulk GaN c -axis LA sound velocity of $8160 \pm 200\ \text{m/s}$, which is close to the recently reported value of $8020\ \text{m/s}$.¹⁵

A comparison experiment in a $5\text{-}\mu\text{m}$ -thick bulk GaN thin film without AlGaIn substrate¹³ shows no acoustic phonon oscillation, indicating the importance of the strain-induced piezoelectric field in the initiation of the coherent oscillation. Similar to our previous experimental results,⁹ the observed coherent acoustic phonons here should also be activated by the carrier-induced piezoelectric field screening in the strained GaN thin film. Thus activated coherent phonon oscillation with nonzero time-dependent displacement would then modulate the strain-induced piezoelectric field and result in the observed probe absorption oscillation. This phenomenon could be described by the frequency domain Franz-Keldysh effect,¹⁶ with a characteristic of absorption modulation sign change around the band gap. By differentiating traces in Figs. 1(a) and 1(b) [shown in inset in Fig. 1(a) as normalized oscillation periods], we found an obvious sign change (or 180° phase change) between these induced oscillations, which is attributed to the Franz-Keldysh effect due to different probing wavelengths. The Franz-Keldysh effect states the relation between probe absorption α and probe energy $\hbar\omega$ when a uniform electric field F is applied in the material with

$$\alpha(\hbar\omega) = \frac{A_0}{2\pi} \left(\frac{2m_r}{\hbar^2} \right)^{3/2} \sqrt{\hbar\theta_F} [-\eta \text{Ai}^2(\eta) + \text{Ai}'^2(\eta)], \quad (1)$$

where $\hbar\theta_F \equiv (\hbar^2 e^2 F^2 / 2m_r)^{1/3}$ and $\eta \equiv (E_g - \hbar\omega) / \hbar\theta_F$, m_r is the reduced effective mass and E_g is the GaN band gap energy. $\text{Ai}(\eta)$ represents the Airy function of variable η and $\text{Ai}'(\eta)$ is its derivative. By plotting the normalized absorption modulation versus pump wavelength with a fixed carrier density of $1 \times 10^{18}\ \text{cm}^{-3}$ in Fig. 3, the zero-crossing wavelength is found to be $\sim 370\ \text{nm}$. This zero-crossing wavelength is close to but larger than the expected GaN band gap value of 362 nm, probably due to existing band tail states that lower the value of η thus resulting a redshift in zero-

FIG. 3. Normalized absorption modulation due to coherent phonon oscillation of the strained GaN. The normalized carrier density is $1 \times 10^{18} \text{ cm}^{-3}$.

crossing probe energy. By comparing the experimental result of Fig. 3 with a theoretical simulation model based on Eq. (1),^{16–19} an order of $10^{-2}\%$ of strain modulation in bulk GaN can be extracted under our experimental condition.

According to fitting, the acoustic oscillation decay time in Fig. 1 is ~ 300 ps, longer than any coherent phonon oscillations reported before. This increase of decay time supports our previous hypothesis that the coherent phonon oscillation decay time in InGaN MQWs was dominated by dephasing between different oscillators instead of acoustic phonon lifetime.⁹ Dephasing in current acoustic phonon oscillation might have several causes; one was due to excitation linewidth. With a 3.6 nm spectral full width at half maximum (FWHM), a thus created self-longitudinal interference pattern would have an uncertainty in its spacing width, with $0.39 \mu\text{m}^{-1}$ spread in phonon wave vector. This would turn out to be negligible compared with another factor: the finite sample thickness. With a total sample thickness $\sim 2.4 \mu\text{m}$, the $1.56 \mu\text{m}^{-1}$ uncertainty in phonon wave vector corresponds to a dephasing time ~ 200 ps,⁹ close to our experimental results.

Our result indicates that the observed short oscillation decay time in previous experiments might be limited by fi-

nite sample width of artificial quantum structures. Bulk materials with larger sample thickness can avoid this disadvantage. With the method of longitudinal interference, our demonstration thus provides the direction toward achieving long-lasting high-frequency coherent acoustic phonons.

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