Ultra-long-lived coherent acoustic phonons in GaN single crystals

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Abstract. We report our experimental and theoretical studies on generation and time-resolved detection of coherent acoustic phonons (CAPs) in bulk GaN single crystals, using the femtosecond optical pump-probe spectroscopy. A train of 100-fs-wide, ultraviolet laser pump pulses with the photon energy far above of the GaN bandgap induced electronic stress at the crystal surface and triggered propagation of CAP oscillations. Subsequently, time-delayed, low-intensity, 100-fs-wide, ultraviolet/infrared (one/two-colour setup) probe pulses monitored CAP propagation either near the surface or deeply inside the GaN crystal. The amplitude of CAP oscillations was on the order of $10^{-5}-10^{-6}$ of the probe-beam transient differential reflectivity signal, and was only dependent on the pump-beam absorption coefficient. The CAP oscillation frequency was dispersionless (proportional to the probe-beam wave vector) with the slope corresponding to 8000 m/s—the speed of sound in GaN. In our two-colour arrangement, we could detect CAP oscillations as deeply as ~0.1 mm inside the crystal (the pump-probe delay ~13.2 ns), since the CAP signal attenuation was only limited by the absorption of the infrared probe light in GaN. The latter indicated that the intrinsic CAP lifetime in our crystals was actually ultra long and was estimated to be of the order of at least 100 ns.

1. Introduction

Acoustic phonon oscillations in solids have been recently investigated using ultrafast optical techniques and have attracted intense interest for their potential in photo-acoustic microscopy of nanostructures, THz acousto-optic modulation, and as coherent sources for phonon spectroscopy [1-4]. In semiconducting materials, coherent acoustic phonons (CAPs) can be excited with ultrashort optical pump pulses via a variety of mechanisms, such as electronic and/or thermal stress [1], deformation potential coupling, or piezoelectric effect. In GaN, first CAP oscillations have been studied by Huang et al. [2], in strained, 0.6-µm-thick films deposited on AlGaN substrates. Recently, we have shown that electronic-stress-induced CAPs could be efficiently generated in GaN crystals using femtosecond ultraviolet, above-bandgap, pump pulses and detected with far-below-bandgap infrared probe photons [5, 6]. Here, we demonstrate that CAPs in GaN bulk samples can propagate macroscopic distances without losing their coherence.

2. GaN crystal growth and femtosecond pump-probe spectroscopy

Our GaN single crystals were grown with a high-pressure, solution-growth method [7]. The growth process was carried out at an external nitrogen gas pressure of 8-14 kbar and in the 1350-1600 °C
temperature range, because of a high, high-temperature solubility of GaN. The crystals exhibited excellent wurtzite crystalline structure and typically formed ~0.5-mm-thick platelets with sizes of up to 3×4 mm².

Figure 1 presents our femtosecond optical pump-probe setup for generation and time-resolved detection of CAPs. Both one-colour and two-colour pump-probe spectroscopy experiments were performed in reflection mode, using a commercial mode-locked Ti:sapphire laser (repetition rate 76 MHz), as well as the homemade third- and second-harmonic generators. The ultraviolet (UV) pulses with the width of ~100 fs and photon energies ranging from 4.71 to 4.42 eV and 3.53 to 3.43 eV, respectively, were used as the pump and focused on the GaN surface with a spot diameter of ~20 μm, the incident angle of ~30º, and the fluence of ~0.04 mJ/cm² per pulse. The probe pulses, used to detect CAPs, were either near-infrared light, directly generated by the Ti:sapphire laser, or UV light after passing the second-harmonic generator. The probe pulse, perpendicular to the sample surface, had the width of ~100 fs and their fluence was always much smaller (at least of the factor of 10) than that of the pump.

We measured the time-resolved normalized change in the reflectivity $\Delta R/R$ of the probe-beam over a time-delay window of ~500 ps with additional fixed delays of up to 13 ns. The UV pump pulses absorbed at the crystal surface generated the thermal and/or electronic stress and triggered a strain transient (CAP pulse), which, while propagating inside the sample, modulated its local index of refraction $n$, leading to the $\Delta R/R$ signal, detected by the delayed probe pulses.

3. Experimental results

3.1. One-colour pump-probe spectroscopy

Figure 2(a) shows the time-resolved $\Delta R/R$ transient, obtained using our one-colour pump-probe setup operating at 355 nm. We note the CAP oscillations, which are superimposed on top of the $\Delta R/R$ electronic (carrier relaxation) signal. To get a clear view of the oscillations, we subtracted the initial pulse rise and the exponential background from the raw $\Delta R/R$ data, as is shown in figure 2(b). The resulting oscillations exhibit a single frequency of 122 GHz [see inset in figure 2(b)], as well as a strong attenuation with a decay time $\tau_d$ of ~70 ps.
Figure 2. $\Delta R/R$ signal measured in one-colour configuration at the wavelength of 355 nm. (a) Full, time-resolved $\Delta R/R$ transient. (b) CAP oscillations extracted from the $\Delta R/R$ signal. The solid line is simple-damped sinusoidal fit, based on equation (1). The inset in (b) is a fast Fourier transform (FFT) of the oscillatory data.

The fit shown in figure 2(b) follows the expression derived by Thomsen et al. [1] and Wu et al. [6]:

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

where the amplitude $A$, and the phase $\varphi$, are the only pump-beam-energy dependent parameters and were introduced by Wu et al. [6]. $f$ is the frequency the CAP oscillations, and, since it comes from the self-inference of the probe light, reflected from the sample at near normal incidence, it is given by [1]

$$
f = \frac{nv}{\pi} k_{\text{probe}} = \frac{2nv}{\lambda_{\text{probe}},}
$$

where $v$ is the speed of sound in GaN and $k_{\text{probe}} = 2\pi/\lambda_{\text{probe}}$ is the probe-light wave vector. Finally, $\tau_d$ is the experimental decay time and

$$
\frac{1}{\tau_d} = \frac{1}{\tau_{\text{phonon}}} + v\alpha_{\text{prb}},
$$

where $\tau_{\text{phonon}}$ is the CAP intrinsic decay time, and $\alpha_{\text{prb}}$ is the probe light absorption coefficient. In one-colour experiments at 355 nm, $1/\tau_d = v\alpha_{\text{prb}}$, making impossible even to estimate the actual $\tau_{\text{phonon}}$.

3.2. Two-colour pump-probe spectroscopy

The two-colour experimental setup was used to investigate the intrinsic lifetime of CAPs. The ~800-nm wavelength of the probe light corresponds to the photon energy of ~1.5 eV, which is far below the GaN bandgap. Thus, in this case, the probe beam experiences only a very small absorption ($\alpha_{\text{prb}} = 50$ cm$^{-1}$) when it travels in our high-quality GaN crystal, which is clearly seen in figure 3. We note that within the 450-ps experimental window [figure 3(a)], we observe essentially non-attenuated CAP signal with $f = 52.1$ GHz [see inset in figure 3(a)], in full agreement with equation (2).
Figure 3. $\Delta R/R$ signals obtained using two-colour spectroscopy. (a) CAP oscillations (dots) and the corresponding FFT plot (inset) within a 450-ps time window. (b) CAP oscillations (dots) measured in several time-delay windows: 0.2-0.35 ns, 1-1.15 ns, 2.5-2.65 ns, and 10-10.15 ns, at the 267-nm pump and the 800-nm probe. The solid lines in (a) and (b) represent the fit based on equation (1).

In order to study CAP propagation at much longer times, we added fixed optical delays between the pump and probe beams so in each case, the translational delay stage could scan an individual, 150-ps-wide time window, as is shown in figure 3(b). We note that the 10-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. The solid lines in figure 3(b), representing the fit based on equation (2) with the single decay constant $\tau_d = 19.2 \pm 0.4$ ns. Since $\alpha_{prb} = 50$ cm$^{-1}$ at 800 nm and for our crystals $v = 8002$ m/s [2, 4], $\alpha_{prb} v \approx 25$ ns, and, therefore, following equation (3), the intrinsic $\tau_{phonon}$ can be estimated to be $\approx 83$ ns at room temperature. The actual $\tau_{phonon}$ value is likely to be even longer (>100 ns), since our 100-fs probe pulses disperse as they propagate inside GaN, what would be another source of attenuation and contribute to $\tau_d$.

4. Conclusion
We have studied generation and propagation of CAP transients in the bulk GaN single crystals using both one- and two-colour femtosecond pump-probe spectroscopy techniques. Using the IR probe beam with the photon energy far below the GaN bandgap and, correspondingly, very low propagation attenuation, we observed the CAP oscillations propagating as deeply, as $\approx 0.1$ mm under the crystal surface with their intrinsic dephasing time to be estimated to be of the order of at least 100 ns. Our future work will focus on a precise determination of $\tau_{phonon}$ using a novel spectroscopy setup with an arbitrary pump-probe delay.

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