Ultrafast Terahertz Nanoseismology of GaInN/GaN Multiple Quantum Wells

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Terahertz (THz) emission spectroscopy and microscopy are applied to investigate the electron and lattice dynamics of Ga$_{0.8}$In$_{0.2}$N/GaN multiple quantum wells (MQWs). The THz emission consists of three distinct, differently timed signals, whose physical mechanisms are attributed to i) laser-induced ultrafast dynamical screening of built-in bias electric field in MQWs followed by ii) capacitive charge oscillation of the excited carriers and iii) the coherent acoustic phonon (CAP)-driven polarization surge at the discontinuity between the GaN capping layer and air. These multifunctional optical responses show strong dependence on the quantum well width and photon energies. The temporal separation between the first and third THz pulses corresponds to the propagation of the CAP across the GaN capping layer of the MQW structure, whose thickness can thus be determined with 10 nm precision.

1. Introduction

Materials subjected to ultrafast optical excitation undergo various types of nonequilibrium dynamics in their electronic, lattice, and spin subsystems. In particular, ultrafast phonon dynamics in functional materials, apart from being of general fundamental research interest, is also important from the point of view of optimized thermal management in devices. The generation of longitudinal coherent acoustic phonons (CAPs) via the ultrafast optical excitation of charge carriers has been explored extensively in different materials and device structures. These include superlattice semiconductor heterostructure thin films,[19] metals,[20] oxides,[21] 2D materials,[22] and multiple quantum wells (MQWs).[5,6] Several possible mechanisms for generating CAPs in the abovementioned materials and device systems include the thermoelastic effect in metals[23] and modulation of the deformation potential and piezoelectric effect in semiconductors.[7–13]

Raman spectroscopy[14,15] is normally employed to study the vibrational dynamics of high-frequency and high-energy optical phonons in materials. However, it is difficult for conventional Raman spectroscopy to detect low-energy CAPs with frequencies in the terahertz (THz) range and below.[16] CAPs can also be measured using X-ray diffraction[17] and time-resolved optical diffraction.[18]

In this work, we apply pump-wavelength-tunable THz emission spectroscopy to study the CAP excitation dynamics in Ga$_{0.8}$In$_{0.2}$N/GaN MQWs. In our samples, the emitted THz waves have several origins: dynamical screening of built-in electric fields and charge oscillation in MQWs, and CAP-to-THz wave conversion at the GaN/air interface. The CAPs are lattice strain waves that are generated via the inverse piezoelectric effect within the photoexcited GaInN/GaN QWs and propagate in the GaN barriers and cap layer of the MQW structure. Both the generation of the CAPs and dynamical screening are strongly dependent on the excitation photon energies for the transitions between 1h–1e and 1h–2e quantum levels within the QWs (see the explanation for the applied selection rules in the following sections).

The choice of GaInN/GaN MQW is owing to the fact that the CAPs generated in GaInN/GaN QWs have significantly higher amplitudes than other piezoelectric materials and heterostructure superlattices[19] while GaN works as an efficient low-loss medium for CAP propagation.[20]

High-quality material growth is required to generate high-amplitude CAPs in semiconductor superlattice
heterostructures.[21] Epitaxial semiconductor crystal growth with high-quality interfaces and layer structures provides conditions for the efficient generation of acoustic phonons.[22] The propagation of CAPs in a piezoelectric medium leads to acoustically generated THz emission at the interface.[23,24] Although Armstrong et al. have reported THz generation at the Al/GaN and AlN/GaN interface,[24] the emission mechanism and propagation properties of acoustic THz phonons, especially those generated within the MQW systems, have not yet been investigated in detail.

Here, we discuss the complex dynamics observed in the optical responses of GaInN/GaN MQWs, resulting in the excitation of ultrafast electronic polarization and lattice dynamics within the sample. Applying THz emission microscopy, we also measured a lateral thickness distribution in GaN cap layer of our samples with 10 nm precision, which proved that scanning laser THz emission microscopy (LTEM) is a highly efficient contactless nanoseismology technique.

2. Samples

The samples used in this experiment (Figure 1, inset) were fabricated using low-pressure metal–organic vapor-phase epitaxy (LP-MOVPE). First, a high-quality 2 µm GaN buffer layer was grown on a 300 µm thick sapphire (Al2O3) substrate, followed by 10 Ga0.8In0.2N QWs of identical widths, sandwiched between 7.2 nm thick GaN barriers. Finally, the MQW structures were encapsulated within a 180 nm thick GaN capping layer. The crystallographic growth direction of all materials was along the c-axis. We prepared three different samples with quantum well widths, \( L_z = 1.5, 2.4, \) and 3.0 nm.

The GaN buffer layer between the substrate and MQW structure accommodates for the difference in lattice parameters between the sapphire substrate and GaN.[25,26]

3. Experimental Section

Here, an LTEM system was used to study the THz emission from GaInN/GaN MQW samples. The experimental configuration details of the LTEM are shown in Figure 1. A Ti:sapphire laser (Spectra-Physics MaiTai HPTK-W) with a pulse width of \( \approx 100 \text{ fs} \) and a repetition rate of 80 MHz was used to excite the MQW samples. The pump wavelengths used in this experiment were chosen such that the photon energy was below the bandgap of the GaN barrier of 3.44 eV, but above the effective bandgap of the Ga0.8In0.2N QWs; thus, the optical transitions only occurred within the quantum wells. The fluence and beam spot diameter used in this experiment were 20 µJ cm\(^{-2}\) and 100 µm, respectively. To match the mentioned photon energy in exciting the MQW region, a wavelength converter (Spectra-Physics GWU-23FSHT-W) was used for the second harmonic of the Ti:sapphire laser (pump beam) generated in a barium borate (BBO) crystal. The laser beam was split into a pump and a variable delay probe beam. The pump beam was modulated using a mechanical optical chopper at a frequency of 2 kHz to facilitate lock-in detection. The THz emission was collected by parabolic mirrors and focused on a dipole-type low-temperature-grown gallium arsenide (LT-GaAs) photoconductive antenna (PCA), which was then gated by the probe beam. Finally, the THz emission signal was retrieved after preamplification and signal processing. The detailed LTEM system design and working principles are reported elsewhere.[27] Figure 2a illustrates the excitation geometry and THz emission from the MQW sample. The internal bias in the MQW sample was directed perpendicular to the surface, which means that the transient dipole vector was also directed normal to the sample surface.[28] Optical excitation \((\hbar \omega)\) and detection were thus performed at an oblique angle of 45°, in order to facilitate the free-space THz generation from these samples.[28]

4. Terahertz Emission Mechanism

The lattice mismatch between Ga0.8In0.2N QW material and GaN barriers produces a permanent strain of 2% inside the QWs, in the growth direction.[13] This strain, together with the piezoelectric nature of GaInN and GaN, gives rise to a strong built-in electric field in the QWs of the order of 3 MV cm\(^{-1}\).[29] which leads to a pronounced quantum-confined Stark effect (QCSE) within the QWs.[30,31] Optical excitation using a femto-second (fs) laser pulse below the bandgap of GaN, but above the effective bandgap of GaInN QWs, excites charge carriers (electrons and holes) in spatially polarized states within the QWs, thus creating an optically induced electric dipole in the GaInN QW, with a polarity opposite to that of the built-in electric field.[32,33]

Thus, intense ultrafast optical excitation of the MQW structure leads to partial or complete screening of the built-in bias field inside the QWs, on the timescale of the optical excitation.[34] This effect, known as coherent dynamical screening effect,[13,35] leads to the following consequences: i) pulsed ultrafast THz emission as a result of ultrafast polarization dynamics within the QWs associated with the fs-timescale screening of
the built-in bias field and ii) simultaneous (partial) release of
the mechanical strain within the QWs, which is no longer bal-
anced by the built-in electric field via the piezoelectric coupling
mechanism. The latter inverse piezoelectric effect generates
THz-frequency CAPs within the MQW structure. Figure 2b
illustrates the band structure modification and generation of
CAPs in an optically excited GaInN/GaN MQW structure. The
theoretical and experimental analyses of the electronic and
acoustic processes involved can be found in refs. [8,9,13,28,32],
while the detailed microscopic theory of CAP generation in
GaInN/GaN systems is presented in ref. [6]. The latter can be
simplified into a loaded string model

\[ \frac{\partial^2 U}{\partial t^2} - c_s^2 \frac{\partial^2 U}{\partial z^2} = S(z,t) \]  

where \( U(z,t) \) corresponds to the amplitude of the acoustic
pulse, \( c_s \) is the speed of sound in the piezoelectric crystal, and
\( S(z,t) \) is the driving function (loading term), which depends on
the photoexcited carriers.[6]

The CAP THz mechanical strain pulse propagating within
a piezoelectric material creates an associated electric polariza-
tion wave packet, propagating through the crystal at a constant
speed of sound. As such, this polarization wave packet does
not lead to electromagnetic THz emission. However, once
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Figure 3 shows a typical detected THz signal from the optically
pumped GaInN/GaN MQW sample. Two pronounced signal
peaks can be distinguished: the first signal at \( t \approx 14.7 \text{ ps} \) (first
peak) labeled as \( T_1 \) and the second signal at \( t \approx 379 \text{ ps} \) (second
peak) labeled as \( T_2 \). The signal at \( T_1 \) is radiated due to
the dynamical screening of the built-in field inside the MQW.[13,28,32]
The signal detected in the THz time waveform at \( T_2 \) (second
signal peak in Figure 3) corresponds to THz emission due to
CAPs, because the transient time \( (\Delta T) \) between the two peaks
\( (\approx 23.2 \text{ ps}) \) corresponds to the propagation time of sound across
the GaN capping layer with a thickness of 185 nm,[24] given the
speed of sound in GaN of 8 nm ps\(^{-1}\).[41]

Figure 4 shows the THz emission waveform recorded over
a time span of 500 ps. The first peak at \( \approx 24.5 \text{ ps} \) corresponds
to dynamical screening to the polarization with the QWs. The
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ciated electric polarization, which serves as the source of the
electromagnetic THz emission into the free space[23,24] (see the
sketch in Figure 2a). In our experiments, CAPs were gener-
ated within the GaInN QWs. The interference of multiple CAP
wave packets generated within individual QWs leads to the pro-
duction of zone-folded CAPs,[13] while the solitary CAP pulse
generated within the QW nearest to the surface propagates
within the GaN cap layer toward the sample surface as a single
acoustic pulse.[13]

The THz emission mechanism from CAPs is as follows:
CAP, generated within the QW nearest to the sample surface,
propagates with the constant speed of sound across the GaN
cap layer and carries the associated electric polarization created
via the piezoelectric mechanism. On reflection at the GaN/air
interface, featuring the maximum discontinuity of the acoustic
impedance, the mechanical strain in the CAP flips its sign,
thus leading to the transient modification of the associated elec-
tric polarization. The latter causes electromagnetic THz emis-
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MQWs. The acoustic pulse propagating within the GaN buffer layer toward the GaN/substrate leads to the THz emission arising at ≈275.6 ps. The inset figures show the magnified version of the main THz time-domain waveforms; the left inset shows the typical THz emission from our MQW sample, and the right inset shows the acoustic pulse arriving at the GaN buffer and sapphire substrate interface.

Figure 5 shows the THz emission signal in the time domain and the respective frequency responses, for three different MQW samples. In the sample with \( L_z = 1.5 \text{ nm} \), the dynamical screening (first peak) arises at \( \approx 11.8 \text{ ps} \), and the acoustic peak (second peak) arises at 36.3 ps, i.e., \( \approx 24.5 \text{ ps} \) later. In the sample with \( L_z = 2.4 \text{ nm} \), the dynamical screening arises at \( \approx 11.8 \text{ ps} \), with the acoustic peak arising at 35.8 ps, i.e., \( \approx 24.0 \text{ ps} \) apart. In the sample with \( L_z = 3.0 \text{ nm} \), the dynamical screening (first peak) arises at \( \approx 11.8 \text{ ps} \), and the acoustic peak arises at 35.3 ps, i.e., \( \approx 23.5 \text{ ps} \) apart. The time stamp of the first peak signal remains nearly the same in all the sample waveforms, whereas the signal due to the acoustic pulse changes both with respect to relative timing and to peak-to-peak amplitude.

The peak-to-peak amplitude of the THz emission due to dynamical screening increases as the quantum well width (\( L_z \)) increases. In contrast, the peak-to-peak amplitude of the THz emission due to the acoustic pulse (second peak) shows an irregular trend and variation in the time stamps. The respective frequency response of THz emission from all three samples, covering the band of 3 THz, is shown in Figure 5b, which is limited by the LT-GaAs dipole photoconductive antenna detection bandwidth and sensitivity.

The magnitude of the THz emission due to dynamical screening depends solely on the quantum well width.[13,28,32] This can be viewed as a process of discharging a nanocapacitor, represented by the strained piezoelectric InGaN QWs containing built-in electric field.[28] The MQW with large \( L_z \) will store more electrostatic energy, increasing the peak-to-peak amplitude of the dynamical screening THz emission as \( L_z \) increases.[28] However, in the case of the acoustic pulse (second peak), the emission magnitude does not solely depend on the built-in constant (2%) strain in the superlattice structure of the MQW sample. It also depends on the CAP wave packet propagation dynamics in GaN, CAPs transient time in capping layer thickness,[24] and specular scattering due to interfaces and surface roughness.[13]

According to the basic relation of piezoelectric constitutive (Equation (2)), the CAPs experience local perturbation of the electric field in piezoelectric materials

\[
T = -cS - eE \quad (2)
\]

where \( T \) is the stress, \( S \) is the strain, \( E \) is the electric field, \( c \) is the coefficient of stiffness, and \( e \) is the piezoelectric coefficient. The perturbation of the electric field of CAP wave packets in piezoelectric materials and the electron–phonon interactions cause attenuation of CAPs.[14] Therefore, the acoustic pulse in Figure 5a exhibits an irregular amplitude trend. Furthermore, the time stamp variation of the acoustic peak is mainly due to the spatial thickness of the capping layer.

Figure 6 shows the power-dependence measurement of the peak-to-peak amplitudes of THz radiation signals with the QW widths of 1.5, 2.4, and 3.0 nm as a function of fluence in the range of 0–20 \( \mu \text{J cm}^{-2} \). Figure 6a shows the trend for dynamical screening THz signal, while Figure 6b shows the CAP-generated THz signal trend. The peak-to-peak THz emission for dynamical screening signal increased linearly for all samples. The THz emission response of the quantum well with \( L_z = 3.0 \text{ nm} \) shows an increase of roughly 20–30% compared with other samples, because the optical absorption in the sample increases as the quantum well width increases, which in return increases the population of excited electron hole pairs leading to strong THz emission.[13] The peak-to-peak emission in the samples with \( L_z = 1.5 \text{ nm} \) and \( L_z = 2.4 \text{ nm} \) shows almost the same trend. As the QW width increased, the THz emission becomes stronger. The linear trend without any saturation in the fluence range of 0–20 \( \mu \text{J cm}^{-2} \) confirms that the total screening of the built-in field in the QWs was not achieved in
the present experiments. It has been shown that the total built-in field screening in our MQW structures is only achieved at a much higher optical fluence of the order of 0.5 mJ cm$^{-2}$.[13,28,32]

In Figure 6b, the power-dependence trend of the CAP-generated THz pulse is shown. All samples showed a small increasing linear trend. The sample with a QW width of 3.0 nm shows a stronger peak-to-peak amplitude in comparison to other samples, while the samples with QW widths of 1.5 and 2.4 nm show almost the same THz emission peak-to-peak amplitude trend. The power-dependence trend for the CAP-generated THz pulse shows the same trend as the dynamical screening THz signal because, during dynamical screening, the impulsive strain changes strongly depend on optically created polarized charge carriers that screen the built-in bias. Therefore, a sample with a larger QW width results in a larger absolute strain release, a higher amplitude for CAPs, and a stronger polarization current spike at the GaN surface. As mentioned above, the pumping fluence range of our experiment is limited and is not sufficient to fully screen the built-in bias, and hence, to release all of the strain within the QWs. This leaves residual strain in the MQW samples during the excitation pulse duration and results in a very weak linear trend for all the samples.

Figure 7 shows the pump wavelength dependence of the THz emission from the MQW samples. The temporal waveforms at different excitation optical wavelengths are shown in Figure 7a, and the magnitude of the multifunctional responses (dynamical screening, oscillatory part, and acoustic pulse) from the time-domain THz waveforms is summarized in Figure 7b.

**Figure 5.** a) Time spectra of THz emission comparison for three different MQW samples, along with b) their frequency response.

**Figure 6.** Power dependence. a) Electric field peak-to-peak for dynamical screening. b) Electric field peak-to-peak for acoustic pulse. The lines are the guide to the eye.
The inset of Figure 7b shows a comparison of the wavelength-dependence measurement between $L_z = 3.0$ nm and $L_z = 1.5$ nm. Excitation photon energies used here are in the range of 2.88–3.44 eV (below the bandgap of GaN and toward the bandgap of Ga$_{0.8}$In$_{0.2}$N). As shown in Figure 7a, once the photon energy starts to increase, such that it shifts toward the bandgap of GaN ($\approx 3.44$ eV), an increase in the peak-to-peak amplitude of the THz emission is observed in terms of multiple signal peaks in the temporal waveforms of THz emission, which includes dynamical screening at 25.0 ps, ripples (oscillatory THz emission part), and acoustically driven THz emission at 47.8 ps. A sharp increase in the THz emission amplitudes is observed until the photon energy of 3.02 eV for the sample with $L_z = 3.0$ nm is reached. After that, the peak-to-peak amplitudes start to decrease as the photon energy increases until the bandgap of GaN (see Figure 7b).

An abrupt increasing trend in THz emission can be seen where the photon energy starts to match the bandgap of GaInN for 1h–2e optical transitions.[36] The strong built-in piezoelectric field along with QCSE in the MQW breaks the symmetry of the potential profile within the QW and modifies the wavefunction overlap of e–h pairs, which assists the 1h–2e transitions (transitions, which are otherwise parity forbidden).[36–38] A stronger overlap of the e–h wavefunctions of 1h–2e with respect to 1h–1e increases the probability of 1h–2e transitions and so eases the parity-forbidden transitions (see Figure 7c). Here, the THz emission is governed by the separation of photoexcited charge carriers in the built-in field; most electrons are excited to the strongly polarized 2e level and thus screen the built-in field. Figure 7c illustrates the overall overlap of the wavefunctions of 1h–2e in comparison to 1h–1e. The measured and calculated values for the 1h–2e transition energy in ref. [36] indicate that transition energies increase as the quantum well width decreases, as shown in Figure 7b (inset), which explains the observations for $L_z = 3.0$ nm and $L_z = 1.5$ nm. Above the energy for the 1h–2e excitation, an abrupt decrease in the THz
emission amplitude was observed, which might be attributed to the decrease in the transition probabilities and/or carrier tunneling to the neighboring QW. Finally, when the photon energy matches the bandgap of GaN (3.44 eV), no prominent THz signals are detected either through dynamical screening or acoustic pulses in the MQW. This THz emission mechanism is defined primarily by the surge current at the GaN surface, which is a typical THz emission mechanism for (bulk) GaN.[39,40]

We now describe the results of the LTEM imaging of the GaInN/GaN MQW samples. The 3.00 mm × 3.50 mm spatial area near the edge of the MQW sample was raster-scanned using an ≈250 µm diameter spot of the pump laser beam. Figure 8a,b shows the LTEM images for dynamical screening and acoustically driven THz emission amplitudes, respectively. This allowed us to estimate the local thickness of the GaN cap layer in our samples. The results of the GaN cap layer thickness distribution imaging in Figure 8c were determined by calculating the GaN layer thickness using the transient time ΔT (the difference between the peak positions of dynamical screening – generated first THz pulse at T₁ and peak position of CAP-generated second THz pulse at T₂) using the known value of the speed of sound in the GaN layer of 8 nm ps⁻¹[41] (see Figure 3). Figure 8d illustrates the overall thickness profile of the GaN capping layer. An almost constant, position-independent timing of THz emission from dynamical screening amplitude is observed, indicating an almost constant MQW thickness across the sample because the THz emission is solely dependent on the average thickness of the QWs (see Figure 5). However, the spatial distribution of the timing of an acoustically driven THz emission shows patches of high- (white), medium- (light blue), and low (dark blue)-amplitude THz emission because the acoustically driven THz emission depends not only on the buried strain in the MQW but also on the acoustic properties of the GaN cap layer. A comparison of the acoustic pulse THz emission with the calculated thickness of the capping layer shows a negative correlation. When the thickness increases, the intensity of the acoustic pulses decreases, and vice versa, because the acoustic wave propagation experiences attenuation and dispersion while traveling through a medium. Here, we report a 10 nm resolution in GaN cap layer thickness measurement, demonstrating that LTEM is a highly efficient contactless nanoseismology technique.

Figure 8. Laser THz emission microscopy (LTEM) imaging for a) dynamical screening, b) acoustic signal, c) capping layer thickness calculation, and d) line profile of capping layer thickness.
6. Conclusion

In conclusion, we report on the simultaneously observed correlated THz emission through the dynamical screening of the internal bias field in strained piezoelectric Ga0.3In0.2N/GaN MQWs, and strain-induced CAP oscillation, which also leads to THz emission, using the LITEM system. The LITEM imaging of the THz emission amplitudes due to the dynamical screening provides the profile of the buried average thickness of the MQW, whereas that of acoustic pulses shows the profile of the GaN capping layer thickness assuming a constant speed of sound in GaN. We observed, for the first time, the THz emission from coherent longitudinal acoustic phonons in MQW samples such that the efficiency of THz emission from CAPs is comparable in strength to that of screening-driven purely electronic THz signals, even though the emission mechanisms are quite different. Our experimental results show a solid prospective for contact-free highly efficient nanoseismology for buried layer structures in semiconductor devices and structures using LITEM.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

coherent acoustic phonons, dynamical screening, GaIn/GaN quantum wells, inverse piezoelectric effect, laser terahertz emission microscopy, nanoseismology, terahertz emission

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