Defect-related temperature dependence of THz emission from GaAs/AlGaAs MQWs grown on off- and on-axis substrates

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Simultaneous molecular beam epitaxial growth of GaAs/AlGaAs multiple quantum wells on two different substrates (one on GaAs (100) and another on a GaAs substrate misoriented by 4° in the (111) direction) resulted in samples of similar structure, but having different defect profiles. The on-axis sample had a higher defect density and more types of electron traps than the off-axis counterpart. Temperature-dependent terahertz (THz) emission and temperature-dependent photoluminescence were measured; in both cases, an intensity quenching was observed between 75 K – 250 K for the on-axis sample, but not in the off-axis sample. We attribute the THz emission quenching to the electron traps present in the sample, which decreases the photocarriers participating in setting up the surface field. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/1.5004597

INTRODUCTION

Defects in bulk semiconductors and heterostructures can have considerable influence on different applications and corresponding operating temperatures. Whether intentionally incorporated or not, defects can affect structure, carrier lifetime, radiative efficiency, and other properties.1,2 As an example, GaAs/AlGaAs quantum wells are archetypal heterostructures which have traditionally been used for infrared laser optoelectronics;3 this requires that the quantum wells have excellent quality interfaces and minimal defect incorporation in order to achieve high radiative efficiency.4,5 Other than the optimization of parameters during the growth process, one technique that has been employed to reduce the defect densities is the use of the slightly misoriented substrates.5–7 High quality GaAs has an EL2 defect concentration of around ~2-40 x1015 cm−3, and lower concentrations (<0.1 x 1015 cm−3) of other common electron traps.8 Conversely, intentionally increasing defects through low-temperature growth or post-growth treatments (where AsGa defect concentrations are at ~1020 cm−3 after growth and ~1017 - 1018 cm−3 after post-growth annealing9), are often done on GaAs in preparation for faster applications such as photoconductive antennas or switches.10,11

A direct means of identifying and measuring defects is through capacitive means such as deep level transient spectroscopy (DLTS).12 It is a destructive technique requiring metal contacts be deposited on the sample to form a Schottky diode, or that the region of interest be part of a p-n junction. It is, however, capable of identifying the defect through the activation energy, identify whether it is a donor or trap, and detect defects in low concentration. But in cases when defect

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identification is not necessary, temperature-dependent photoluminescence (PL) is a nondestructive, alternative means of obtaining defect-related information for direct gap semiconductors. Some shallow defects may present emission peaks at low temperatures, and deeper defects such as carrier traps can compete with the radiative recombination at higher temperatures.\textsuperscript{1,2}

Meanwhile, terahertz (THz) spectroscopy is fast becoming a standard technique in characterizing semiconductors.\textsuperscript{13} The widely-used THz time-domain spectroscopy can be a tool in obtaining physical material information, as well as ultrafast carrier dynamics.\textsuperscript{14–16} GaAs-based heterostructures are also finding applications in the THz field as many groups are working on developing THz sources and detectors based on a QCL design;\textsuperscript{17,18} the operating temperatures vary greatly among the developing devices. In terms of material characterization, a few studies have been conducted on the temperature-dependence of bulk semiconductor surfaces.\textsuperscript{19–22} However, no universal behavior has been found for THz emission intensity with temperature, as each differs from one sample to the other. GaAs and InSb were reported to have increasing intensities with decreasing temperature,\textsuperscript{19} InP and InMnAs experiencing reversal of THz polarization with temperature,\textsuperscript{21,22} and InAs having no temperature dependence.\textsuperscript{20}

In this study, we measured the temperature-dependent THz emission and temperature-dependent PL of high quality GaAs/AlGaAs multiple quantum wells (MQW) grown on two different substrates – GaAs (100) and GaAs (100) with a 4° misorientation in the (111) direction. Results show that the on-axis grown sample exhibited defect-related PL quenching, as well as THz emission-quenching within the same temperature range. Although THz emission and radiative recombination occur on different timescales and arise from different mechanisms, results show that the presence of defects can have considerable influence on both these processes. The correlation between the PL and THz emission of our samples show that MQW THz emission is sensitive to the presence of defects. While defects can compete with the radiative recombination process, defects can also decrease the THz emission at lowered temperatures due to the active electron traps which localize the photocarriers, decreasing the surge current.

**METHODOLOGY**

GaAs/AlGaAs quantum wells were grown simultaneously using a Riber 32P MBE machine, albeit on separate n\textsuperscript{+} GaAs substrates, one on-axis (100) and one off-axis (4° towards the (111) direction). Figure 1 shows a layer schematic for the epitaxial layers. The substrate temperatures were kept between 580-630°C, with a V/III flux ratio of 16/1. Samples consist of 1 \(\mu\)m-thick GaAs buffer, 40 periods of 100 Å AlGaAs barrier and 50 Å undoped GaAs wells. The samples were

![Layer schematic for the epitaxially grown samples. The 500 \(\mu\)m-thick substrates are placed side by side, and the layers are then grown simultaneously.](https://example.com/layer_schematic.png)

**FIG. 1.** Left: Layer schematic for the epitaxially grown samples. The 500 \(\mu\)m-thick substrates are placed side by side, and the layers are then grown simultaneously. Right: Illustration of energy band diagram showing radiative and non-radiative recombination pathways for photo-excited electrons.
capped with 100 Å-thick Si-doped GaAs. The samples are simply labeled as “On-axis” and “Off-axis” for identification. The periodicities of the QWs were calculated from X-ray diffraction (XRD) rocking curves, using a high resolution double-crystal diffractometer. The samples were scanned at (002) orientation with a step size of 0.0007°. An illustration of the possible pathways by which photocarriers exit the conduction band are shown in Fig. 1 (right), including the radiative band-to-band transitions (PL) and the non-radiative electron capture by identified traps.

Photoluminescence was performed using a continuous-wave Ar⁺ ion laser (λ = 488 nm, beam diameter = 0.5 mm), with an average power of 3 mW. PL was collected using appropriate optics, focused onto a spectrometer, and detected by a GaAs photomultiplier tube. To conduct temperature-dependent measurements, the sample was placed on a cold finger inside a closed-cycle helium cryostat equipped with a temperature controller.

In order to examine the presence of carrier traps, the samples underwent deep level transient spectroscopy. The samples were etched into circular mesas and contacts were deposited via thermal evaporation. The results of the DLTS measurements were reported in a previous work but will be summarized in the succeeding discussion.

Carrier lifetime measurements were carried out using a double optical pump THz time-domain emission spectrometer (DOP-THz-TDES). Figure 2 shows the schematic diagrams of the THz spectrometers used in this study. The DOP THz-TDES set-up (Fig. 2, left) is similar to the standard THz TDES spectrometer (Fig. 2, right), modified by splitting the optical pump pulse into two cross-polarized, 800 nm wavelength beams. One beam functions as a ‘THz generation pump’ (s-polarized, 30 mW) while the other passes through a delay stage and functions as a ‘carrier injection pump’ (p-polarized, 60 mW), effectively time-resolving the set-up. The optical beams are spatially overlapped on the sample at 45° to the surface normal (beam diameters ∼800 µm). The ‘THz generation pump’ sets up the electric field responsible for the THz emission, and the THz delay line is fixed at the maximum. The ‘carrier injection pump’ delay line is scanned; upon arrival on the sample surface, the injected carriers cause optical screening, manifested as a sudden decrease in THz emission intensity. As the injected carriers relax, the emission intensity returns to its initial value since the THz delay line is fixed at the emission peak. The time constant, therefore, reflects the population decay of the injected free carriers.

Terahertz emission from the MQWs was measured in a separate standard emission-reflection geometry THz-TDES set-up (Fig. 1, right) with the sample kept inside a cryogenic chamber evacuated by a closed-cycle He cryostat. The excitation was provided by a Ti:Sapphire laser, emitting 100-fs pulses at a wavelength of 800 nm. The excitation beam was split equally into a pump and a probe beam. The p-polarized pump beam was mechanically chopped at a frequency of 2 kHz, and focused onto the sample surface (beam diameter ∼60 µm), incident at 45°, with an average power of ∼60 mW. The THz radiation emitted by the sample was collected and focused onto a detector through a pair
of off-axis parabolic mirrors. A low temperature grown GaAs (LT-GaAs) photoconductive dipole antenna was used as a detector, optically triggered by the time-delayed probe beam.

RESULTS AND DISCUSSION

In order to establish the quality and uniformity of the MBE-grown samples, several room temperature measurements were conducted – XRD rocking curves for the period thickness, PL to confirm well widths and radiative efficiency, and carrier lifetime measurement. Results from the high resolution XRD rocking curves, showing only AlGaAs satellite peaks, are presented in Fig. 3 (left) (a logarithmic y-axis scale was used in order to discern the satellite peaks more conveniently). The average period thicknesses are calculated to be 172±5 Å and 167±5 Å for the on-axis and off-axis samples, respectively. Correspondingly, the PL spectra are shown in Fig. 3 (right). A clear separation of the C1-HH1 (1.519 eV) and C1-LH1 (1.542 eV) transitions is observed and the calculated well-widths obtained from energy peak positions using the effective mass method (finite-well model) are in agreement with the nominal growth value. No significant differences in energy peak positions nor FWHM is observed, showing the uniformity in growth conditions and average sample well-widths. The PL peak positions indicate that the difference in periodicity observed in the XRD diffraction should likely be accounted for by difference in AlGaAs barrier thickness and not due to the GaAs well thickness. Additionally, the 10 K PL is shown as an inset in Fig. 5 showing increased radiative efficiency, much narrower linewidth. At T = 10 K, only the C1-HH1 recombination pathway is active, and no defect level radiation is detected.

Carrier lifetime was measured using the DOP-THz-TDES set-up. The technique of using three synchronized optical pulses to measure carrier lifetime have previously been applied by other groups to electrically biased semiconductor layers and devices. By applying this technique to bare semiconductor surfaces (unbiased), rather than the externally applied field, the electric field set-up by the THz generation pump is probed by the carrier injection pump instead. The most similar in alignment to the system used in this study in that of Loata, et al., with the exception of the applied bias. The DOP-THz-TDES emission decay curves are presented in Fig. 4. The kinks appearing at the initial transition are artefacts arising from the slight differences in the two THz TDS waveforms at time delays before the main peak. The decay constant was obtained from the slope of linear fits to the semilogarithmic plots (not shown). The room temperature carrier lifetimes are found to be ~3.8 ns and ~4.4 ns for the on-axis and off-axis samples, respectively. The long carrier lifetimes, as well as the structural confirmation of nominal values show good quality samples were grown on either substrate. The samples are similar in structure, but exhibit slight differences in radiative efficiency and carrier lifetime at room temperature.

DLTS measurements were previously obtained for the samples, as well as the consequent effect on the temperature behavior on PL. Results have shown that the samples are high quality, having low to very low trap concentration. For the on-axis grown sample, electron traps such as EL2 (1.2 x 10¹⁵ cm⁻³), EL6 (2 x 10¹⁴ cm⁻³), and EL10 (3 x 10¹⁴ cm⁻³) were identified. And for the off-axis

FIG. 3. Left: HRXRD rocking curves showing AlGaAs satellite peaks; Right: room temperature photoluminescence spectra, inset shows the normalized PL to show uniformity in well-widths.
FIG. 4. Carrier lifetimes at room temperature, measured using DOP THz TDES technique, the on-axis sample shows slightly faster decay than the off-axis sample. Offsets are provided for a clearer view of the plots.

sample, only an unidentified deep electron trap with a -1.09 eV activation energy was identified, and at a much lower density of \(3 \times 10^{13} \text{ cm}^{-3}\).

One common practice when studying the onset of non-radiative processes is by plotting the PL intensity against reciprocal temperature. The shape of the curve is usually an elbow-like transition, and has been described by a phenomenological expression where an Arrhenius-type activation energy can be obtained. Figure 5 shows the Arrhenius-type plot of the integrated PL intensity of the samples with reciprocal temperature (a nonlinear temperature scale is shown on the top of the figure for guidance). The off-axis sample exhibits the expected PL behavior,[29,30] with the activation of non-radiative processes beginning at \(\sim 100 \text{ K}\). For the on-axis sample, however, a drastic PL quenching is observed between temperatures of 80 K – 200 K. This PL quenching is attributed to the presence of easily-accessible traps.[23] The presence of these carrier traps contribute to non-radiative recombination pathways for the carriers, and thus compete with the PL process.

THz emission from the MQW samples was also measured at different temperatures. At this point, it should be noted that the THz emission from these MQW samples have been previously shown to originate primarily (>85%) from the MQW layers, rather than the underlying GaAs buffer,[31] owing to the enhanced absorption of quantum wells relative to bulk. Figure 6 shows representative plots at 300 K, 75 K and 10 K. At any given temperature, the THz emission from the off-axis sample
FIG. 6. Representative THz emission spectra at 10 K, 75 K and 300 K. Left: THz emission waveforms. Right: THz emission power spectra (FFT). Offsets have been provided for ease of viewing.

was greater in intensity. Small differences in the tail of the lineshape are observed (Fig. 6, left). The corresponding THz power spectra (Fig. 6, right) shows similar bandwidths for both samples, with some difference shape in the low frequency side. More noticeable is the difference between their emission intensity. Figure 7 (inset) shows the temperature behavior of the integrated THz power (area under curve of the frequency spectrum). A similar Arrhenius-type plot is shown in Fig. 7, obtained by taking the integrated THz power and plotting against reciprocal temperature (1/T). Going from high to low temperature, the off-axis sample’s THz emission is seen to increase slightly; and from 200 K downwards, the intensity tapers off. For the on-axis sample, the THz intensity continually decreases.

FIG. 7. Temperature dependence of the integrated THz power in an Arrhenius-type plot similar to the PL. (The inset shows the normal THz power versus temperature plot.) The on-axis grown sample shows THz intensity quenching, in the same temperature region (shaded area) as that for PL quenching.
as temperature is lowered further. Interestingly, a THz emission intensity quenching is also observed below 200 K and the signal recovers at 75 K; with the lowest intensity found at 100 K. Although the THz intensity decrease is not comparable to the PL intensity quenching, quite noticeably, the two phenomena occur in the roughly same temperature region. In both Arrhenius-type plots, no quenching was exhibited by the off-axis sample. One difference between the temperature-dependence of the PL and the THz, is that the PL quenching recovers below 80 K.

As mentioned earlier, previous reports on the temperature behavior of THz emission from bulk semiconductors differ from sample to sample.\textsuperscript{19–22} For bulk GaAs, the report by B. B. Hu, et al\textsuperscript{19} presents increasing THz emission with decreasing temperature, which the authors attribute to the increasing mobility. However, other semiconductors behaved differently, such as the non-dependence to temperature by n-InMnAs\textsuperscript{21} above 100 K, or the polarity reversal found for InMnAs\textsuperscript{21} and InP.\textsuperscript{22} This implies that temperature-dependence of THz emission is more complex, depending on each sample’s THz generation mechanism.

In bulk GaAs, the THz emission originates from a combination of the photo-Dember current, and a surge current arising from surface states, with the latter being more dominant at optical pump energies close to the bandgap.\textsuperscript{16} The surge current that forms upon photoexcitation is proportional to the photocarrier concentration. However, the presence of deep levels in the samples causes localization of the trapped electrons.\textsuperscript{1,32,33} This depletes the photocarrier population participating in the creation of the drift (and diffusion) currents. As temperature is decreased, deep levels are filled. In the case of photoluminescence, the trapping of carriers (in this case, electrons) is a straightforward competition to the radiative process especially at temperatures where other scattering processes are also active. As temperature is further decreased, phonon-mediated scattering events decrease, and the radiative recombination becomes much more efficient as the overlap between electron and hole wavefunctions increase.\textsuperscript{32} These greatly increase the photoluminescence. The measured THz emission on the other hand, is not subject to the same crystal momentum selection rules as PL, and does not rely on electron-hole wavefunction overlap. Thus it is more representative of the overall photocarrier population. Furthermore, although radiative recombination efficiency increases as temperature is decreased, it does not necessarily imply that there are more carriers overall. Rather, instead of losing carriers to scattering, they are able to recombine radiatively, instead. Thus, localized (trapped) electrons decrease THz emission because they are unable to participate in setting up the surge current. The compromised electron population of the on-axis grown sample is emphasized by its low THz emission intensity, relative to the less defective off-axis sample. Even though the on-axis sample’s radiative recombination efficiency (PL) is higher at low temperature, the THz emission hints at carrier localization. We note that THz emission intensity has been shown to decrease with decreasing growth temperature, where more defects are expected to form.\textsuperscript{24}

Although the two processes originate from different mechanisms, the temperature-dependent MQW THz emission mimicking the PL intensity quenching supports the premise that the THz emission is sensitive to the presence of defects in these high quality samples. The reason as to why the quenching occurs in the region is yet difficult to determine, since the exact location of the traps are unknown and it is difficult to isolate the contributions of the different types of defects found. Although in other reports, the EL2 trap has been known to transition into a metastable state referred to as EL2\textsuperscript{a} causing photocurrent quenching, albeit in a different temperature region (80-125 K).\textsuperscript{34} A recent report by Reshchikov\textsuperscript{33} explores the PL quenching by multi-center defect levels causing redirection from radiative acceptor paths to nonradiative defects by using a phenomenological model for several III-V and II-VI semiconductors.

**CONCLUSIONS**

In this study, two MQWs simultaneously grown on differently oriented substrates were shown to be structurally similar from the XRD and room temperature PL. The two samples used have been previously characterized using DLTS and reported to have different defect profiles. The temperature dependence of their PL and THz emission were measured. The on-axis grown sample which contains more electron traps, exhibited PL intensity quenching, as well as decreased THz emission intensity as temperature is decreased. The off-axis grown sample which has a lower trap density, exhibited
the usual PL temperature-dependence, but showed reduction in THz emission below 200 K. The correlation between the temperature behaviors of PL and THz emission show that THz emission spectroscopy is sensitive to the presence of deep level defects. The decrease in THz emission at low temperature is attributed to carrier localization as the electron traps fill.

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