Characterizing Bandgap Gratings in GaAs : AlAs Superlattice Structures Using Interface Phonons

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***Abstract*—Interface Raman modes were used to study quantum-well intermixing in GaAs : AlAs superlattice (SL) structures using room-temperature spatially resolved Raman spectroscopy. The intermixing was observed to degrade the interface mode intensity, which can be used as a sensitive indicator of the SL quality. This feature, along with spatially resolved photoluminescence, was used to investigate bandgap modulation in periodically intermixed bandgap gratings, fabricated using implantation induced disordering. Using interface modes instead of bulk-like modes is a promising avenue for characterizing SL structures that rely on intricate bandgap features.**

***Index Terms*—Gratings, intermixing semiconductor superlattices (SLs), phase matching, Raman spectroscopy.**

QUANTUM-WELL-INTERMIXING (QWI) technologies have evolved to become a strong candidate for the realization of photonic integrated circuits. These technologies are now widely applied for the fabrication of devices such as optical switching matrices and for laser diodes integrated with passive sections such as gratings and external cavities [1]. It has been demonstrated that QWI enables the tailoring of the bandgap, and hence the linear and nonlinear optical coefficients of these structures. Of particular relevance to this work, modulation of the second-order susceptibility coefficient $\chi^{(2)}$ in periodically intermixed superlattice (SL) has been used to demonstrate quasi-phase-matching (QPM). To achieve QPM via this technique at wavelengths around 1.55 μm, first-order bandgap gratings with periods on the order of 3 μm are necessary. The principle has recently been implemented in GaAs : AlAs SL structures [2], however, the experimentally measured QPM efficiency was lower than predictions. This is most likely due to limitations in the QWI technology used, the scattering due to the unavoidable refractive index grating generated along with the bandgap grating, and fabrication imperfections.

A nondestructive characterization technique is needed to aid in the optimization and design of these bandgap gratings. Optical measurements, including spatially resolved photoluminescence (PL), are commonly used to observe the bandgap modulation. However, the PL resolution is intrinsically limited by the diffusion length of the excited carriers, which is $\sim$1 μm in GaAs [3]. Consequently, there is a lower limit on the size of the period which can be studied and on the sharpness of the interface between different regions that can be detected. Sharp modulation between the different bandgap regions has been shown to be important to achieve efficient QPM. For this reason, characterization by Raman spectroscopy offers an important advantage over PL. The Raman signal is generated only from the directly illuminated portion of the sample, and so the spatial resolution is limited only by the laser spot size. One of the important QWI attributes is interface and transition region quality, and so for modulated structures with feature sizes on the order of 1–2 μm or smaller, this superior resolution can be crucial.

Besides offering improved spatial resolution, Raman spectroscopy also provides information that cannot be obtained from PL. Shifts in Raman mode wavenumbers, peak widths, and changes in the relative strengths can indicate variations in lattice order, stress, and can be used for chemical profiling [4]. Further, quantum-confined heterostructures, specifically SL structures, have unique phonon modes that appear in the Raman spectra. These have been thoroughly studied for GaAs : AlAs SLs and include zone-folded acoustic phonons, confined optical phonons (COPs), and interface phonon (IF) modes [5]. In this work, spatially resolved Raman spectroscopy is used to study bandgap gratings fabricated in GaAs : AlAs SL structures using ion implantation-induced disordering, with a specific focus on IF Raman modes.

The wafer structure used in this work is comprised of a 600-nm 14 : 14 GaAs : AlAs SL. The SL core layer is placed between a 1.1-μm upper cladding and a lower cladding consisting of 4.3-μm $Al_{0.6}Ga_{0.4}As$. The details of the structure are published in [2]. To avoid absorption in the GaAs cap and improve the signal-to-noise ratio (SNR) of the Raman signal, we probe the sample edge-on with the beam centered on the SL core. Selection rules dictate that the phonons observed in this configuration have wavevector components parallel to the interfaces and are referred to as transverse optic (TO) modes.

Three samples originating from the same GaAs : AlAs SL structure were systematically examined. First, the as-grown SL (Sample 1) was measured and then the spectra are compared with spectra from a fully intermixed SL (Sample 2), where the SL core is expected to have been converted to $Al_{0.5}Ga_{0.5}As$. Sample 2 was intermixed using high-energy ion implantation and subsequent rapid thermal annealing at 850 °C for 30 s. The implanted ions were 4-MeV As at a dose of 2×10^{13} ions/cm^2. Finally, we refer to these two endpoints as we investigate a periodically intermixed sample, produced using a similar ion-implantation process as Sample 2 and a periodic mask produced from a 1.5-μm gold layer.

Measurements were carried out at room temperature with a Jobin-Yvon Horiba micro-Raman system using a CW 532-nm laser. The Raman measurement parameters were 1.8-mW power focused through a 100$\times$ objective, a 100-μm confocal hole, and 5-s accumulation time. The PL was measured with a power of

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TABLE I
OBSERVED RAMAN PEAKS FOR THE SL CORE SAMPLES

Sample	Peak Designation	Position (cm^{-1})	Expected Position[a] (cm^{-1})
SL layer in as-grown sample	GaAs-TO [IF]	267 [278, 284]	268 [277, 282, 284][b]
	AlAs-TO	360	360.1
	AlAs-IF/LO	380.5	382.5[c]
SL layer in intermixed sample	GaAs-like TO	260.4	260.8 (50 % Al)
	AlAs-like TO	359.4	360.0 (50 % Al)
	AlAs-like LO	383.6	385.9 (50 % Al)
$Al_{0.6}Ga_{0.4}As$ cladding	GaAs-like TO	259.1	259.5
	AlAs-like TO	360.4	360.1
	AlAs-like LO	386.6	389.6

[a]Fit to empirical equations in [11] except, [b]Calculation with continuum model of [10] and [c]Continuum model of [12].

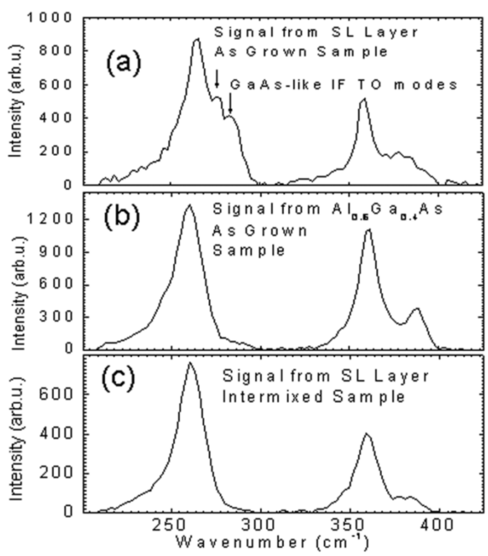

Fig. 1. Raman spectra from (a) the SL layer and (b) the as-grown $Al_{0.6}Ga_{0.4}As$ cladding, and (c) the SL layer in the intermixed sample.

0.18 mW. The results from large area reference samples, namely Samples 1 and 2, will be discussed first. The peaks appearing in the Raman spectra are compiled in Table I, while representative spectra from the SL layer in the as-grown sample, the SL layer in the intermixed sample, and the $Al_{0.6}Ga_{0.4}As$ cladding in the as-grown sample are shown in Fig. 1. The room-temperature PL wavelength generated by the SL was 752 nm for TE polarization, and 732 for TM. These values match those predicted by the band structure calculations of [6]. Also seen on the samples is a peak at 612 nm corresponding to the signal from $Al_{0.6}Ga_{0.4}As$.

The similarity between Raman spectra of the intermixed SL and the $Al_{0.6}Ga_{0.4}As$ alloy is consistent with the expectation that the fully intermixed SL will approximately be $Al_{0.5}Ga_{0.5}As$ alloy. The spectrum of the as-grown SL, however, shows a considerable shift of the lowest frequency peak from about 260 cm^{-1} on the alloy to 267 cm^{-1} on the intact SL. These peaks are identified, respectively, as the GaAs-like TO alloy mode and the bulk GaAs-TO mode originating in the GaAs layers of the SL. Evidence of confined TO modes on the as-grown SL, which would appear below the GaAs-TO peak, were not observed. However, the calculated dispersion along the $\langle 100 \rangle$ to $\langle 110 \rangle$ direction is very limited for this mode [7], and so the confined modes should be very closely spaced after Brillouin zone (BZ) folding. Applying a linear chain model [8] to the 14 : 14 SL, 14 modes arise between the bulk GaAs-TO mode and 256 cm^{-1}, with a maximum mode spacing of 1.2 cm^{-1}. The predicted intensities also drop quickly with increasing order. As such, it is not surprising that the COPs are not resolved.

The mode appearing at 360 cm^{-1} is identified as the AlAs-TO mode on the as-grown SL, and as the AlAs-like TO alloy mode on the intermixed SL and $Al_{0.6}Ga_{0.4}As$ alloy. These modes feature almost no dispersion across the BZ [8], or with varying Al content in the alloy [5], and so no significant shift in this peak is expected or seen. However, the peak width is noticeably smaller on the as-grown SL, consistent with the expectation of greater order in the SL layers than in the alloys.

The highest frequency peak is identified as the AlAs-like LO mode from the $Al_{0.6}Ga_{0.4}As$ layer, and as a broad feature relating to the AlAs-IF modes on the as-grown SL. This feature has been observed in previous SL studies [9], and corresponds to the predictions of a continuum model we have used to predict the Raman modes in the SL [10]. On the intermixed SL, expected to be $Al_{0.5}Ga_{0.5}As$, the peak appears 3 cm^{-1} lower than on the $Al_{0.6}Ga_{0.4}As$ alloy, which compares well with a 4-cm^{-1} shift predicted between these Al fractions by the empirical equations of [11]. As such, we tentatively identify this mode with the $Al_{0.5}Ga_{0.5}As$ AlAs-like LO mode.

The most interesting feature of the SL Raman spectra, when compared with the alloy, is the appearance of the two peaks directly above the GaAs-TO mode. These peaks at 278 and 284 cm^{-1} correspond to the GaAs IF modes as predicted by dielectric [12] and phenomenological continuum models [10], and previously observed in Raman scattering studies [9]. Their presence is a clear indication of the presence of intact SL. For off-resonance Raman measurement, the IF modes are only selected with a probe aligned to an in-plane SL direction.

Further examining the results of the intermixed sample, it is clear to see that the 6-cm^{-1} shift of the GaAs-like TO mode is much larger than that exhibited by the two AlAs-like modes. However, the appearance of the GaAs-IF modes only for the as-grown sample indicates that they will be the strongest gauge of intact SL structure quality, as can be seen in Fig. 1.

A bandgap grating was created through QWI using ion implantation-induced intermixing and subsequent annealing as discussed above. The grating was designed to have a period of 3.8 μm with a 50% duty cycle, although ion straggle and defect diffusion will cause a variation on this duty cycle. The laser spot had a diameter of $\sim$0.5 μm and was centered on the SL layer, where it scanned across several cycles of the grating with a step of 0.1 μm. The incident light was TE polarized which provides optimal coupling with the induced electric field in the SL structure, which is oriented parallel to the growth axis (perpendicular to the beam). The TO modes are essentially oscillating dipoles which align to this field, and so TE polarization provided the best SNR for Raman measurement. The resulting spectra were curve fitted and a collection of modes similar to those found in Samples 1 and 2 were obtained. However, for Sample 3, the modes are periodically modulated as a result of the QWI. The shift in position of the GaAs-like TO peak corresponds to that anticipated from the SL intermixing, providing a strong contrast between intermixed and intermixing suppressed regions. The peak value on the intermixing suppressed region corresponds closely to the expected bulk GaAs-like TO peak at 268 cm^{-1}, and the peak on the intermixed region matches that expected for $Al_{0.5}Ga_{0.5}As$ at 260.8 cm^{-1}[11]. This is a clear indication of

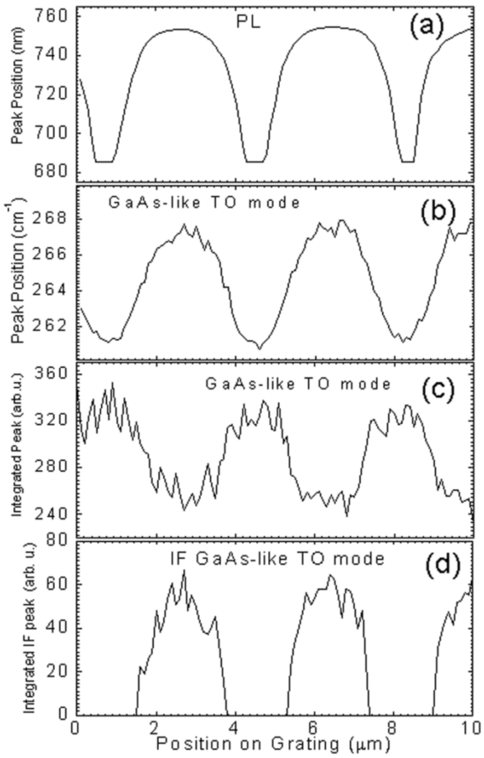

Fig. 2. Profiles along the SL bandgap grating are shown, revealing the (a) PL peak position, (b) GaAs-like TO mode position, (c) GaAs-like TO mode integrated area, and (d) GaAs-like IF integrated area.

the degree of intermixing in both regions of the grating, which in this case alternate between intact SL and the equivalent bulk AlGaAs. The AlAs-like TO and LO modes were detected, but due to their limited shift with Al composition and poor SNR, respectively, their modulation was much less pronounced, being on the order of 1 cm^{-1} between the two different regions. In Fig. 2(b) and (c), the wavenumber and area under the peak for the GaAs-like TO mode are plotted as a function of position along the bandgap grating. In addition, Fig. 2(d) shows the area underneath the GaAs IF mode. It is clear that the peaks are not resolved where the sample is intermixed, and thus provide a superior SNR for detecting SL intermixing in comparison to evidence provided by the GaAs-like TO mode.

The corresponding scan of the PL is provided in Fig. 2(a). The PL peak takes on a value of 757 nm on the intact SL region, compared with 752 nm on the as-grown SL. In the center of the intermixed region, the PL peak is at 682 nm. This compares poorly with the 598-nm peak seen on Sample 2, and suggests incomplete intermixing of the SL in the intermixed part of the grating. The absence of the 598-nm peak may also arise despite the presence of a limited region with this bandgap, due to photo-generated carrier diffusion to a region with the smaller 682-nm bandgap. The PL scan shows clear evidence of lateral photo-generated carrier diffusion, as the regions of smaller bandgap appear larger than those of higher bandgap. This gives the false impression of an asymmetric duty cycle between the intermixed and intermixing-suppressed regions. The Raman profiles give a closer correspondence to the structure since Raman does not rely on carriers. From Fig. 2(b)–(d), it can be seen that the duty cycle of the grating is close to 50% as designed. Inconsistency seems to arise when Raman spectra in which no IF modes appear are correlated with the 682-nm PL peak, which suggests incomplete intermixing. The answer relies in the fact that not detecting the IF modes only suggests that their signal is smaller than the tail of the GaAs-like TO mode along which they appear. However, the results demonstrate the benefits of using IF modes which are bound to provide substantial advantages over the bulk Raman modes which were used in previous work, by us and other groups, to detect lattice damage and intermixing [13].

In summary, GaAs-IF modes were used to characterize intermixing of GaAs : AlAs SL structures through spatially resolved Raman spectroscopy. The intermixing was observed to degrade the IF modes, which were used to investigate the bandgap modulation in a grating fabricated by QWI. Due to the large effect of QWI on the IF modes, using these instead of bulk-like modes is a promising avenue for characterizing intricate features defined by any QWI process. While the technique can also be used for InP compounds, their Raman scattering is weaker than that of GaAs compounds.

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