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To cite this article: Matasit Chikumpa *et al* 2020 *Mater. Res. Express* **7** 105007

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PAPER

OPEN ACCESS

RECEIVED

1 August 2020

REVISED

30 September 2020

ACCEPTED FOR PUBLICATION

2 October 2020

PUBLISHED

14 October 2020

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Raman peak shifts by applied magnetic field in InSb/Al_xIn_{1-x}Sb superlattices

Matasit Chikumpa¹, Zon¹ , Supachok Thainoi¹, Suwit Kiravittaya² , Aniwat Tandraechanurat³, Noppadon Nuntawong⁴, Suwat Sopitpan⁴, Visittapong Yordsri⁴, Chanchana Thanachayanont⁴, Songphol Kanjanachuchai¹ , Somchai Ratanathamphan¹ and Somsak Panyakeow¹

¹ Semiconductor Devices Research Laboratory, Department of Electrical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok, Thailand

² Advanced Optical Technology Laboratory, Department of Electrical and Computer Engineering, Faculty of Engineering, Naresuan University, Phitsanulok, Thailand

³ International School of Engineering (ISE), Faculty of Engineering, Chulalongkorn University, Bangkok, Thailand

⁴ National Science and Technology Development Agency (NSTDA), Pathumthani 12120, Thailand

E-mail: s_panyakeow@yahoo.com

Keywords: InSb/AlInSb, superlattices, molecular beam epitaxy, Raman spectroscopy, magnetic effect

Abstract

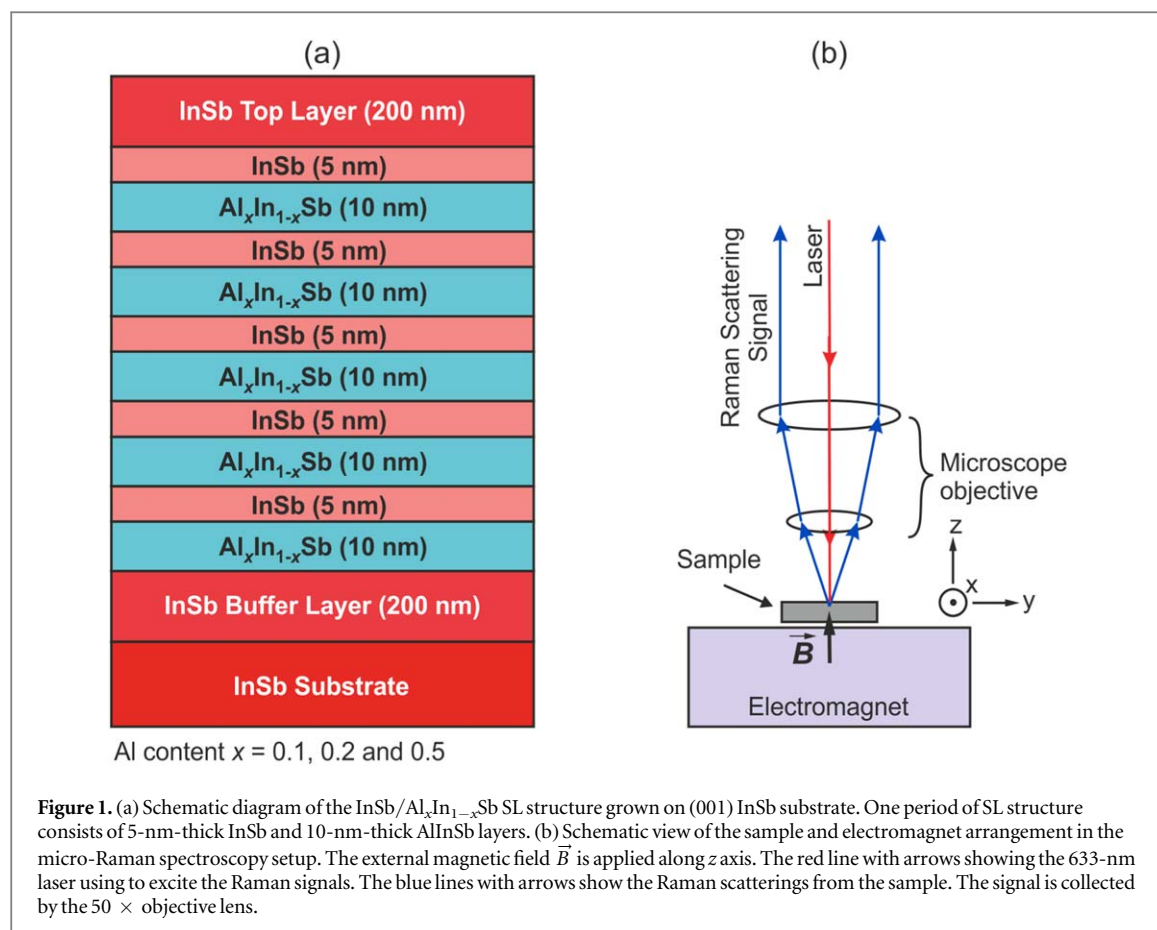
InSb/Al_xIn_{1-x}Sb superlattices (SLs) are grown by molecular beam epitaxy on (001) InSb substrate and Raman scattering spectroscopy of the samples under magnetic field is investigated. Al contents in AlInSb of the samples are varied. All samples are characterized by atomic force microscopy (AFM), X-ray diffraction and Raman scattering spectroscopy. The Raman spectroscopy is done by using excitation laser with 633 nm wavelength and 2 μm beam spot under applied magnetic field from 0 to 170 mT. Both TO and LO Raman peaks from InSb are detected from all samples. There are Raman peak shift of both TO and LO by applied magnetic field. Stronger magnetic effect is found in LO than TO phonon modes. We attribute this effect to the symmetry breaking of the InSb/AlInSb interfaces since the observed roughness of the top InSb layer can qualitatively correlate with the shift.

1. Introduction

In the last decades, III-V compound superlattices (SLs) and quantum nanostructures have been widely investigated as they are promising structures for novel electronic and optoelectronic device applications [1, 2]. Among them, antimonide-based material systems namely InSb, GaSb, AlSb, InGaSb, AlGaSb have been explored for their basic properties of various substrates (GaAs, InAs, GaSb, and InSb) [3–14]. Structural characteristics of both nearly lattice-matched such as InAs/GaSb and many mismatched systems are reported along with their fabrication details as they are the prerequisites for the successful realization of novel nano-devices.

Concerning the device applications of InSb-based systems, InSb/AlInSb SLs or quantum nanostructures can be utilized for realizing high performance optoelectronic devices operating in infrared wavelength range [8, 9, 15–22]. It has also been demonstrated that InSb can be used as Hall bar for magnetic field sensing since the room temperature (RT) electron mobility in InSb is highest among all III-V compounds [23–25]. Typically, two-dimensional electron gas is formed in the high-electron mobility structure based on this material. However, the magneto-optical properties of InSb/AlInSb SLs in visible range have not been well investigated.

Recently, we have observed a small but noticeable Raman scattering peak shifts of samples containing nanostructures [9]. In another work [26], the Raman peak shift is observed in free-standing InSb nanowire sample. The observed results for InSb nanowires [26] and nano-strips [9] are less obvious as compared to other material systems [27, 28]. We thus investigate the InSb/AlInSb SLs. In this work, we report on the realization and Raman peak shifts of InSb/Al_xIn_{1-x}Sb SLs. The Al content x is varied and the shifts of both LO and TO peaks are observed in all Al-contained samples. We have qualitatively explained the origin of these shifts to the



interface inhomogeneity of the InSb/AlInSb interface as the roughness of InSb top layer can be correlated to the interface.

2. Sample preparation

InSb/Al_xIn_{1-x}Sb $\times 5$ SL samples are grown on (001) InSb substrates by molecular beam epitaxy (MBE RIBER Compact 21TM) equipped with an antimony (Sb) valved cracker cell. The In and Al sources are conventional effusion cells. The overall growth process was monitored *in situ* by reflection high energy electron diffraction (RHEED) observation. Prior to the growth, the substrate is pre-heated at 200 °C for 1 h in the preheating chamber. After preheating, it is transferred into the MBE growth chamber and the de-oxidation process is performed. By RHEED observation, the surface is de-oxidized at 380 °C. To minimize the surface roughness after de-oxidation process, 200 nm thick InSb buffer layer with In growth rate of 0.12 monolayer per second (ML/s) is grown on the InSb substrate at de-oxidized temperature 380 °C. Then, Al_xIn_{1-x}Sb (10 nm) layer is grown and followed by InSb (5 nm) layer for complete one period of InSb/AlInSb SL. The Al content x in AlInSb is varied from 0.1 to 0.2 and 0.5 adjusted by the temperature of Al effusion cell in the range of ~ 1010 to ~ 1120 °C for the targeted amount of Al composition. The SL active region is composed of five periods-InSb (5 nm)/AlInSb (10 nm). Finally, 200-nm-thick InSb layer is grown at the topmost of the SL structure as the capping layer. The substrate temperature and In growth rate are fixed at 380 °C, 0.12 ML s⁻¹ for overall MBE growth process. The schematic diagram of InSb/AlInSb SL structure is shown in figure 1(a).

The surface morphology of the grown samples is characterized by the atomic force microscopy (AFM, Seiko SPA-400) in dynamic force mode in air. Crystalline quality, layer thickness and the lattice parameter variation relating to III-V compound composition are probed by an X-ray diffractometer (HR-XRD Rigaku TTRAX III) operated at 50 kV. The Raman spectroscopy (Reinshaw inViaTM) is performed by employing 633-nm excitation laser at RT. Figure 1(b) shows the arrangement of the sample and electromagnet in the micro-Raman spectroscopy setup. In order to investigate the effect of magnetic field on the Raman spectra, the external magnetic field \vec{B} is increased from 0 to 32, 64, 89, 122, 144 and 170 millitesla (mT). The magnetic field is controlled by the applied voltage to an electromagnet. Calibration with a commercial magnetometer is performed before and after the experiment. The magnetic field \vec{B} is applied in z direction.

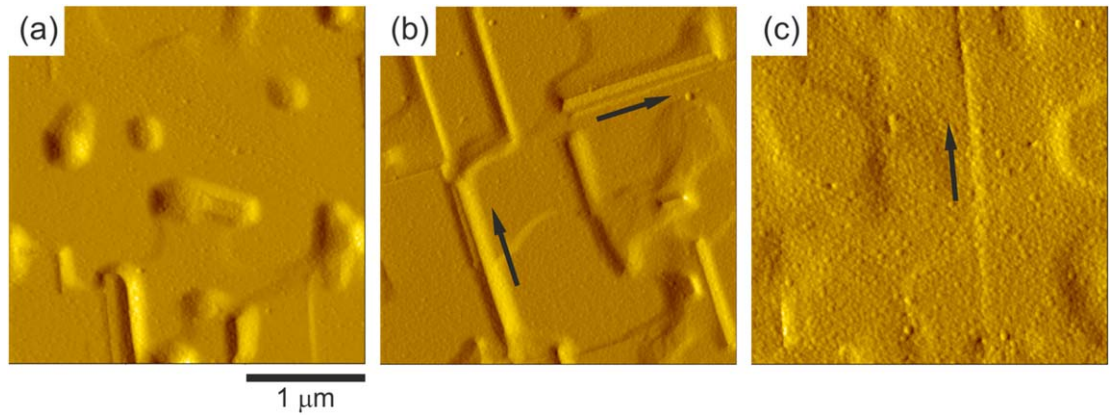


Figure 2. $3 \times 3 \mu\text{m}^2$ AFM images of top InSb layers grown on InSb/ $\text{Al}_x\text{In}_{1-x}\text{Sb}$ SL having Al contents x of (a) 0.1, (b) 0.2 and (c) 0.5. The arrows show the elongated trends observing on the surface.

3. Results and discussion

The surface morphology of the topmost InSb layer is observed by AFM. Figures 2(a)–(c) shows the typical InSb surfaces from the samples having Al content $x = 0.1, 0.2$, and 0.5 . Holes and elongated trends are observed on the surface of the sample with $x = 0.1$. The similar trends get longer and show densely on the $x = 0.2$ sample. The roughness of $x = 0.5$ sample gets higher showing corrugated and stepped surface. The RMS roughness values of the top InSb surfaces of the samples with $x = 0.1, 0.2$ and 0.5 are $1.33, 2.09$ and 2.42 nm , respectively, by probing the $1 \times 1 \mu\text{m}^2$ surface area. This roughness can be related to the interface roughness of underneath InSb/ AlInSb SLs and it might relate to the degree of Raman peak shifts, which are shown below.

HR-XRD analysis is carried out to investigate the crystalline quality, layer thickness and III-V compound composition in the InSb/ $\text{Al}_x\text{In}_{1-x}\text{Sb}$ SL structure. The samples are diffracted by x-ray beam with wavelength of 1.5406 \AA ($\text{Cu K}\alpha_1$) at 0.02° step. Figure 3(a) shows the overall XRD patterns in log (normalized) scale of the three samples with various x and InSb substrate (reference) at the diffraction angle 2θ range between 20° and 80° . InSb peaks from (200) reflection at $2\theta \sim 28^\circ$ and (400) reflection at $2\theta \sim 57^\circ$ are observed [11, 17]. Magnified view of XRD patterns are shown in figure 3(b). The (400) reflection peak of InSb is observed at $2\theta \sim 56.79^\circ$. The reflections at $2\theta \sim 57^\circ\text{--}59^\circ$ can be assigned to the central diffraction peaks from mixtures of epitaxial InSb and $\text{Al}_x\text{In}_{1-x}\text{Sb}$ layers. The diffraction angle 2θ calculation is done by using typical Bragg Law. To evaluate the behaviour of material composition in the investigated SLs, we calculate the diffraction angle 2θ of SL in two models; one for AlInSb peak and one for InSb/ AlInSb mixture peak. The calculated and experimental results are described in table 1. The experimental results of central 2θ reflections are extracted by the function of Lorentz fit. The result from XRD shows monotonic shift and broadening of the InSb/ $\text{Al}_x\text{In}_{1-x}\text{Sb}$ peak when the aluminium content increases (from 0.1 to 0.5). The former is due to the increase of the Al content x while the latter is from the roughness increment. This deliberate growth of SL with low structural quality (high roughness) is done in order to probe the magnetic field induced Raman peak shift, which is shown below.

The Raman spectroscopy is performed to study the strain characteristics of the InSb/ AlInSb SL structure, and the relation between the magnetic field B and the Raman frequency shift. To investigate the effect of external applied B on the Raman spectrum, B is increased from 0 to $32, 64, 89, 122, 144$ and 170 mT while the excitation laser wavelength is fixed at 633 nm . B is aligned along in the z direction. The beam spot size of excitation laser is $\sim 2 \mu\text{m}$ by employing the $50 \times$ objective lens, and the signal is scanned with the acquisition time of 20 s . Figure 4(a) shows the Raman scattering spectrum of SL ($x = 0.1$) measured without B . The three peaks corresponding to the Sb cluster, first and second order InSb are observed at $\sim 140, 170\text{--}200$ and $370\text{--}385 \text{ cm}^{-1}$, respectively [9, 11–14, 29]. Since the vicinity of laser beam spot is $\sim 2 \mu\text{m}$, B interacting with laser beam excitation is limited. With limited area of interaction, however, the evolution of the Raman shift as the function of B is occurred in all samples as shown in figures 4(b)–(d). The strong first order InSb peak is focussed to study the Raman peak shift. When B increases from 0 to 170 mT , the blue-shift of transverse optical (TO) and longitudinal optical (LO) phonon peaks of InSb is observed. The broad Raman scattering features at lower values of B is the resultants from the contributing of TO and LO phonon peaks, and the LO phonon peak is getting stronger and clearly split from TO mode at higher values of B . We speculate that the lattice vibration or polarization of LO phonons is more strongly affected by the external magnetic field than that of TO phonons.

Raman frequency shift as the function of B is summarized as shown in figure 5 for the investigated samples. The peak-positions of LO and TO phonon modes are extracted by fitting with the Gaussian function. The values

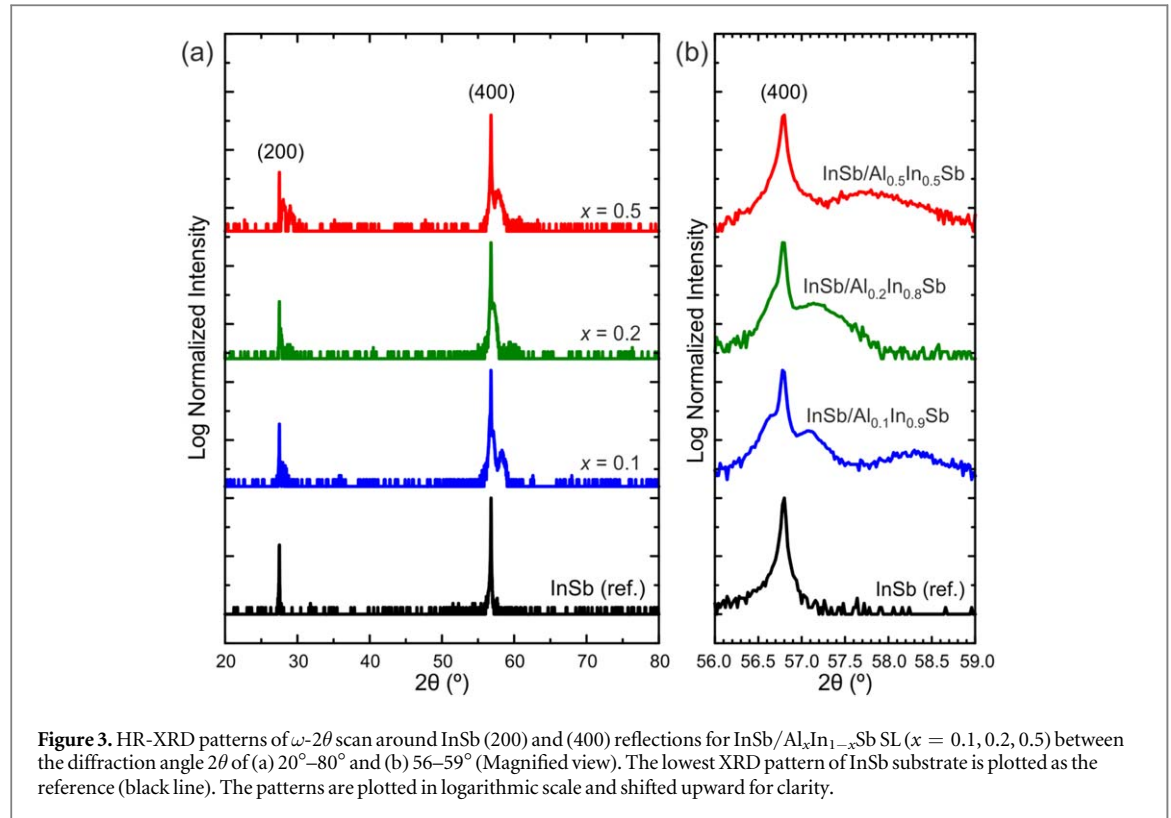


Figure 3. HR-XRD patterns of ω - 2θ scan around InSb (200) and (400) reflections for InSb/Al_xIn_{1-x}Sb SL ($x = 0.1, 0.2, 0.5$) between the diffraction angle 2θ of (a) 20° – 80° and (b) 56 – 59° (Magnified view). The lowest XRD pattern of InSb substrate is plotted as the reference (black line). The patterns are plotted in logarithmic scale and shifted upward for clarity.

Table 1. Comparison of calculated and experimental X-ray diffraction angle 2θ of InSb/Al_xIn_{1-x}Sb superlattices with different Al content x .

Al content x	Calculation						Experiment
	Al _{x} In _{1-x} Sb peak			InSb/Al _{x} In _{1-x} Sb peak			
	$2\theta_-$ (°)	$2\theta_0$ (°)	$2\theta_+$ (°)	$2\theta_-$ (°)	$2\theta_0$ (°)	$2\theta_+$ (°)	
0.1	56.45	57.12	57.79	56.34	57.01	57.68	57.12
0.2	56.78	57.45	58.13	56.56	57.23	57.90	57.23
0.5	57.81	58.48	29.16	57.23	57.90	58.58	57.85

$2\theta_-$ = first satellite peak at negative angle.

$2\theta_0$ = central diffraction angle.

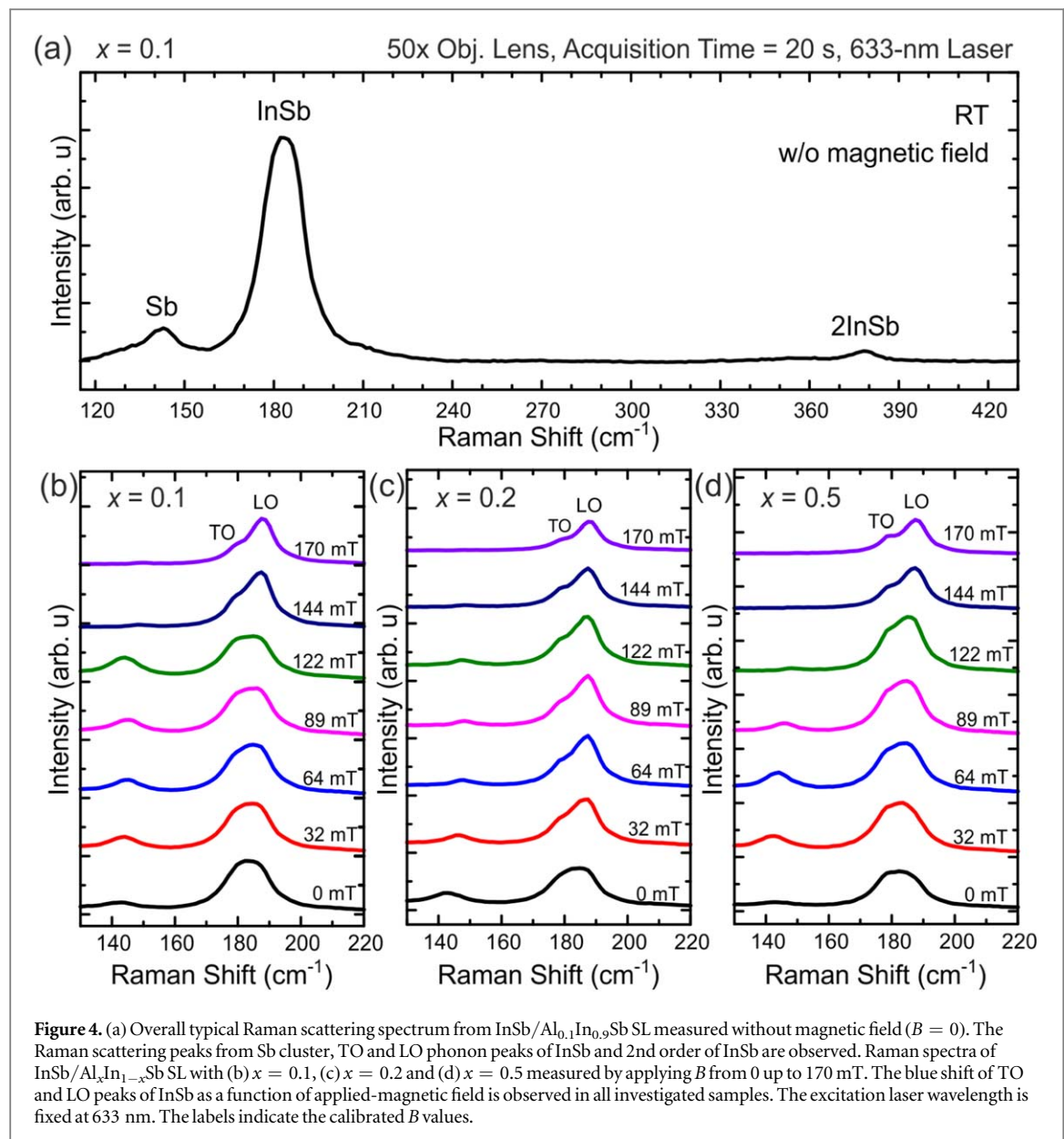
$2\theta_+$ = first satellite peak.

of (LO and TO) phonon peak shifts for InSb/AlInSb SLs with $x = 0.1, 0.2$ and 0.5 are (2.19, 1.59), (2.95, 2.24), and (4.16, 2.03) cm^{-1} , respectively. The slope of shifting data indicates the sensitivity of magnetic effect of each sample. The samples with $x = 0.1$ and 0.2 show similar results with nearly the same slope and discrepancy among the LO and TO lines. The general concept of the Raman frequency shift of the optical phonons, $\Delta\omega$, by the external applied magnetic field, B , can be simply expressed as

$$\Delta\omega = \gamma B,$$

where, γ is an empirical constant (in the unit of cm^{-1}/mT). The experimental results indicate that the γ_{LO} is larger at SL with $x = 0.5$, which can be related to the different lattice vibration due to the lattice parameter variation by increasing x . In this work, the origin of these shifts can be qualitatively explained as the interface inhomogeneity of the InSb/AlInSb interface correlating to the surface corrugation of InSb top layer. Since perfect non-magnetic crystals such as III-V compound semiconductors do not show a magnetic field induced Raman shift [30]. Further theoretical considerations as well as explicit atomistic calculations are still needed to proceed for developing a complete understanding of this phenomenon [30, 31].

Comparison on the Raman peak positions of the three SL samples with various x at $B = 0$ up to 170 mT, the red-shift of the Raman scatterings is revealed as x increases. The fitted TO and LO peak positions of the three SLs with $x = 0.1, 0.2$ and 0.5 at $B = 0$ mT are obtained at (178.75, 185.79), (176.82, 185.05) and (176.63, 183.50) cm^{-1} , respectively. The TO and LO peaks are shifted by 1.93 cm^{-1} and 0.74 cm^{-1} when x increases from 0.1 to 0.2, and 0.19 cm^{-1} and 1.55 cm^{-1} when x increases from 0.2 to 0.5, respectively. The possible explanation for



these shifts is that the dislocations generated by the relaxation of misfit strain are eliminated at the interfaces in the SL. The nearby areas have both the compressive and tensile strained regions. In our case, the red-shift of the Raman phonon peaks can be attributed to the induced tensile strain at the InSb/AlInSb interfaces manipulated by the Al composition in the ternary compound AlInSb.

4. Summary

InSb/ $\text{Al}_x\text{In}_{1-x}\text{Sb}$ SLs with various Al contents ($x = 0.1, 0.2$ and 0.5) have been prepared on InSb substrates by MBE. The surface corrugation of top InSb layer is qualitatively analysed by AFM. The sample with $x = 0.5$ gets most roughening surface among the investigated samples. XRD measurement exhibits the reflections from InSb and the InSb/ $\text{Al}_x\text{In}_{1-x}\text{Sb}$ mixture. By comparing with the calculated values, the material intermixing occurs at the interfaces of SL, and the diffraction peaks are mainly observed from the InSb/ $\text{Al}_x\text{In}_{1-x}\text{Sb}$ mixture. Micro-Raman spectroscopy accompanying with external applied magnetic field B reveals that the influence of magnetic field on the InSb/ $\text{Al}_x\text{In}_{1-x}\text{Sb}$ SL structure. InSb/ $\text{Al}_{0.5}\text{In}_{0.5}\text{Sb}$ SL has the highest magnetic response of phonons to the applied B . The clear blue-shift of TO and LO Raman phonon scatterings of SL is observed when $B = 0$ up to 170 mT. This can be implied to the dissymmetry behaviour of the InSb/AlInSb interfaces which is correlated to the surface roughness of the top InSb layer. Moreover, the red-shifts of TO and LO Raman phonon peaks reveal the relaxation of tensile strain at the interfaces induced by higher Al composition.

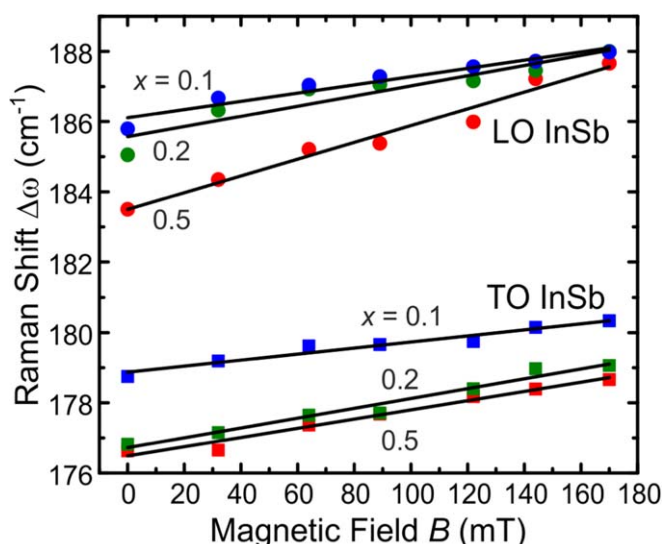


Figure 5. The linear fitting curves of the LO and TO peak shifts $\Delta\omega$ by varying applied magnetic field \vec{B} . The Raman scattering frequencies as a function of \vec{B} for InSb/ $\text{Al}_x\text{In}_{1-x}\text{Sb}$ SL with $x = 0.1, 0.2$ and 0.5 are represented by blue, green and red dots.

Acknowledgments

This research was financially supported by the Research Chair Grant, the National Science and Technology Development Agency (NSTDA), Thailand (Contract No. FDA-CO-2558-1407-TH), the Asian Office of Aerospace Research and Development (AOARD) Grant, co-funded with the Office of Naval Research Global (ONRG) under Grant No. FA 2386-16-1-4003, Thailand Research Fund (Contract No. DPG5380002), NANOTEC, NSTDA, Thailand (Contract No. RES-50-016-21-016), and Chulalongkorn University. Ms. Zon acknowledges support from the Ratchadaphiseksomphot Fund for Postdoctoral Fellowships of Chulalongkorn University.

ORCID iDs

Zon <https://orcid.org/0000-0003-1998-6482>

Suwit Kiravittaya <https://orcid.org/0000-0003-0609-5445>

Songphol Kanjanachuchai <https://orcid.org/0000-0003-4622-4176>

Somsak Panyakeow <https://orcid.org/0000-0002-8618-1839>

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