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Mid-infrared to ultraviolet optical properties of InSb grown on GaAs by molecular beam epitaxy

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Spectroscopic ellipsometry was used to investigate the optical properties of an InSb film grown on a GaAs (100) substrate, and to compare the optical properties of InSb film with those of bulk InSb. The film was grown by molecular beam epitaxy under conditions intended to form 90° misfit dislocations at the InSb-GaAs interface. The complex dielectric function obtained in a wide spectroscopic range from 0.06–4.6 eV shows the critical point transitions E_0 , E_1 , $E_1 + \Delta_1$, E'_0 , and E_2 . The amplitudes, energy transitions, broadenings, and phase angles have been determined using a derivative analysis. Comparing film and bulk critical point results reveal that the epitaxial film is nearly relaxed and has bulk-like optical characteristics. © 2015 AIP Publishing LLC.

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I. INTRODUCTION

InSb offers the highest electron and hole mobilities among group III–V semiconductors. It also possesses a small bandgap E_0 of ~0.2 eV, which makes it suitable for mid-infrared optical devices. These unique properties make InSb highly desirable for integrating with Si for future optical and electronic devices. Since GaAs is nearly lattice-matched to Ge, integration of GaAs with Ge-buffered Si is possible and has been reported.^{1–4} Growth of high quality InSb films on GaAs is challenging due to large lattice mismatch of about ~14.6%. One approach to overcome mismatched epitaxy is to relieve strain energy through the formation of an interfacial misfit (IMF) dislocation array.^{5–7} IMF is attractive because the misfit dislocations (90°) are confined to the film-substrate interface and the subsequent growth follows a layer-by-layer strain-free epitaxy. IMF has been observed in a variety of III–V materials deposited on GaAs (001) substrate under specific growth conditions when it is energetically favorable to form 90° misfit dislocations instead of 60° misfit dislocations.⁸

Existence of 90° misfit dislocation array was reported for the growth of InSb on GaAs (001).^{9,10} The InSb films produced by Williams *et al.*⁹ showed threading dislocations and stacking faults in the film in addition to the 90° misfit dislocations at the interface. Kiely *et al.*¹⁰ fabricated higher quality InSb films and also carried out a detailed study of the interface by cross-sectional transmission electron microscopy (XTEM). The spacing between the misfit dislocations was found to have an average value of ~3 nm. This value nearly matches the expected periodicity of ~3.1 nm for pure 90° misfit dislocations at the InSb/GaAs. The theoretical misfit spacing value for pure 60° misfit dislocations is ~1.6 nm. However, the bulk of the InSb film still showed

defects such as dislocation tangles and threading dislocations in addition to the presence of a rough interface with a ripple period of 40–50 nm. Their work suggests that there is some room for improvement of material quality of InSb film grown on GaAs.

The optical and electronic properties of InSb films formed by IMF need to be evaluated to assess their suitability for various device applications, in particular, around the bandgap region which lies in the mid-infrared energy range. The dielectric response from mid-infrared to ultraviolet and the critical points E_0 , E_1 , $E_1 + \Delta_1$, E'_0 and E_2 in crystalline InSb can be investigated using spectroscopic ellipsometry. The critical parameters include amplitudes, broadening parameters, phase angles, and energy transitions which provide valuable information about the electronic bandstructure of a material, and can be obtained using a derivative analysis of the complex dielectric function.^{11–13}

There are reports on the determination of visible-UV dielectric function and critical point parameters associated with E_1 , $E_1 + \Delta_1$, E'_0 and E_2 in bulk InSb^{11–17} and in InSb film on sapphire.¹⁸ These include even a temperature dependent investigation of E_1 , $E_1 + \Delta_1$, E'_0 and E_2 critical points in bulk InSb.^{11,13} Spectroscopic ellipsometric investigation of the infrared dielectric function around E_0 as well as the critical point parameters has not been reported for bulk samples. A systematic study of critical point parameters in InSb film grown on GaAs has not been reported.

In this paper, we investigate the electronic bandstructure of InSb grown on GaAs using spectroscopic ellipsometry from 0.06 to 4.6 eV at room temperature. The growth of InSb on GaAs by molecular beam epitaxy (MBE) is optimized using IMF arrays. The amplitudes, energy transitions, broadenings, and phase angles are obtained for the critical points E_0 , E_1 , $E_1 + \Delta_1$, E'_0 and E_2 using a derivative analysis. The critical point parameters obtained for the film are in good agreement with the bulk values indicating a crystalline and nearly relaxed film.

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II. GROWTH DETAILS AND STRUCTURAL CHARACTERIZATION

Heteroepitaxial InSb layer was grown on a (001) semi-insulating GaAs substrate using a solid source MBE equipped with arsenic (As) and antimony (Sb) valved cracker sources. The cracker temperature of the Sb source was set at 900 °C to produce predominately Sb and Sb₂ fluxes.¹⁹ The GaAs substrate was pre-heated to 580 °C to remove the native oxide. This was followed by the deposition of a 100 nm-thick GaAs buffer at 580 °C to smoothen the surface. The substrate temperature was then lowered to 540 °C under the As overpressure and annealed at the same temperature for 600 s without As overpressure. Subsequently, the substrate temperature was reduced to 310 °C and the wafer was exposed to Sb flux for 240 s. The annealing of the wafer at 540 °C and its exposure to Sb flux for 240 s are found to be critical for the formation of a single monolayer of Sb atoms on the surface of the substrate prior to InSb growth. It minimizes the formation of 60° misfit dislocation at the surface.⁶ A ~300 nm thick InSb layer was subsequently grown by opening the indium shutter.

The structural quality of the sample was investigated by high-resolution X-ray diffractometry (HR-XRD) and XTEM. Figure 1(a) shows the (004) ω -2 θ curve for the InSb film. The peak at lower ω angle corresponds to InSb layer. The full-width-half-maximum (FWHM) of InSb film peak is ~185 arcseconds. Such small values of FWHM are observed typically only in films at least a micrometer thick.²⁰ Reduction of FWHM in micrometer thick films is attributed to improvement in crystalline quality, particularly in the top portion of the film that is far from the highly defective film-substrate interface. The out-of-plane lattice constant of InSb film was obtained from the symmetric (004) ω -2 θ curve. ω -2 θ curves corresponding to asymmetric reflections (115)

and ($\bar{1}\bar{1}5$) were acquired to obtain the in-plane lattice constant. The in-plane strain $\varepsilon_{\parallel}$ in the InSb film is ~-0.1% indicating that the film is nearly relaxed (~99.3%). The lattice mismatch between the InSb film and GaAs substrate is relieved through the formation of misfit dislocation array at the interface. The XTEM image of the sample as shown in Fig. 1(b) indicates the presence of an IMF layer. The misfit spacing is found to be ~3.2 nm, which nearly matches the theoretical value of ~3.1 nm. The threading dislocation density in the film estimated from TEM images is $<10^8 \text{ cm}^{-2}$. In addition, atomic force microscopy (AFM) showed that the root mean square (RMS) roughness of InSb film is ~1 nm.

III. ELLIPSOMETRY AND CRITICAL POINT ANALYSIS PROCEDURES

J. A. Woollam's infrared variable angle spectroscopic ellipsometer (IR-VASE) and a variable angle spectroscopic ellipsometer (VASE) were used for acquisition of ellipsometric angles Ψ and Δ . The data were acquired at two angles of incidence, 65° and 75°, from 480 to 3630 by 8 cm⁻¹ (0.06 to 0.45 eV) and 1.2 to 4.6 by 0.01 eV on the IR-VASE and VASE, respectively. A reference InSb substrate was characterized along with the InSb film grown on GaAs. Fig. 2(a) shows the experimental and the model pseudo-dielectric function for InSb film sample in the IR energy range. Pseudo-dielectric function denotes the dielectric response of the entire sample (overlayer-film-substrate). The visible-UV pseudo-dielectric function for InSb film is shown in Fig. 3(a). E_1 , $E_1 + \Delta_1$, E'_0 , and E_2 are observed in the visible-UV pseudo-dielectric function. E_0 is masked by the presence of interference oscillations in the infrared range. The feature around 1.5 eV in the visible-UV dielectric function also corresponds to an interference oscillation.

To further investigate the optical response of InSb layer in detail, the film sample was modeled as a three-layer structure consisting of a bottom-most GaAs substrate, InSb film, and a

FIG. 1. (a) HRXRD (004) ω -2 θ scan for an InSb film grown on GaAs (001). (b) XTEM image for the InSb sample showing the misfit dislocations at the interface between InSb and GaAs. The misfit dislocations appear as black dots in the XTEM image, and the spacing between them is estimated to be ~3.2 nm.

FIG. 2. (a) Experimental and model infrared pseudo-dielectric functions for the sample structure consisting of InSb film grown on GaAs (100). The experimental pseudo-dielectric function was obtained from Ψ and Δ acquired at an angle of incidence of 65°. (b) Experimental and model infrared dielectric functions of the InSb film. Experimental data are plotted using dots, and the model data are represented by solid lines.

FIG. 3. (a) Experimental and model visible-UV pseudo-dielectric functions for the sample structure consisting of InSb film grown on GaAs (100). The experimental pseudo-dielectric function was obtained from Ψ and Δ acquired at an angle of incidence of 65° . (b) Experimental and model visible-UV dielectric functions of the InSb film. Experimental data are plotted using dots, and the model data are represented by solid lines.

surface layer. The surface film was modeled as an InSb-oxide layer in the visible-UV range.²¹ It was described as a rough layer in the infrared range where the oxide optical constants are not available. The dielectric function of GaAs substrate used in the modeling was obtained by characterizing it separately. The visible-UV complex dielectric function of InSb film is modeled using four, Kramers-Kronig consistent, parametric oscillators²² which describe the four critical point transitions E_1 , $E_1 + \Delta_1$, E'_0 , and E_2 . The mid-infrared data were analyzed separately using a model containing an E_0 critical point structure. The overlayer thickness (oxide or roughness), the InSb film thickness, and the InSb parametric optical constants were obtained simultaneously using standard ellipsometric data fitting procedures. With the layer thicknesses determined, dielectric function of InSb film was then extracted on a wavelength-by-wavelength basis.²³ The dielectric function obtained in this manner is termed as point-by-point dielectric function. For the bulk InSb sample, essentially the same procedure was used to obtain parametric and point-by-point dielectric functions, using just an overlayer-substrate model.

Point-by-point and model infrared dielectric functions obtained for the InSb film are shown in Fig. 2(b). The direct bandgap E_0 is now seen in the imaginary part of infrared dielectric function, revealing its square root dependence on photon energy. Fig. 3(b) shows point-by-point and model dielectric functions in the visible-UV range. The excellent agreement between the point-by-point and model dielectric function confirms the Kramers-Kronig consistency of the point-by-point function. The amplitude of E_2 peak in the dielectric function of InSb film is ~ 21.8 at 3.87 eV, which is close to the value of 22.0 obtained by Bermudez and Ritz¹⁵ for bulk InSb, whose surface was cleaved and maintained in ultra-high vacuum. Using chemical etching, Aspnes and Studna¹² achieved a value of 21.3 for the amplitude of E_2 peak.

Point-by-point dielectric function is preferred to parameterized dielectric function for numerical derivative analysis.²³ Although the oscillator model provides a very good fit to the raw data, small discrepancies may result near the critical points which can be amplified while calculating

derivatives of the parameterized dielectric function. The visible-UV dielectric function with an energy step size of 0.01 eV is differentiated using 13 Savitzky-Golay smoothing coefficients for second-order derivatives with a polynomial of order 5. The derivatives of the experimental dielectric function were fitted using an analytic expression²³

$$\frac{d^2 \epsilon_{\text{visible-UV}}}{dE^2} = \sum_j \frac{A_j e^{i\Phi_j}}{[E - E_j + i\Gamma_j]^2}, \quad (1)$$

where A_j is the amplitude for transition j , Φ_j is the phase angle, E_j is the critical point energy and Γ_j is the broadening parameter. The summation covers the four transitions E_1 , $E_1 + \Delta_1$, E'_0 , and E_2 . Although numerical differentiation of model is desirable in addition to numerical differentiation of experimental data,²⁴ we ensured that the number of smoothing coefficients was carefully chosen to enhance the signal to noise ratio without distorting the sharp features in experimental data relative to unsmoothed data.

First-order numerical derivatives are used in the determination of direct bandgap E_0 of InSb. The mid-infrared data with 1 meV energy step size were smoothed using 13 Savitzky-Golay coefficients corresponding to a polynomial of order 4. The first derivative of infrared dielectric function was fitted using an analytic lineshape given by²³

$$\frac{d\epsilon_{\text{IR}}}{dE} = \frac{A_{\text{pole}}}{E^3} + \frac{A_{E_0} e^{i\Phi_{E_0}}}{(E - E_0 + i\Gamma_{E_0})^{1/2}}, \quad (2)$$

where A_{E_0} is the amplitude, Φ_{E_0} is the phase angle, and Γ_{E_0} is the broadening parameter for E_0 critical point. A_{pole} is the magnitude of the low-energy pole, which accounts for absorption outside of the infrared range.

The derivatives of the imaginary and real parts of the dielectric function were fitted simultaneously by a least-square procedure which uses the Levenberg-Marquardt algorithm.²⁵ The critical point analysis for an InSb film is shown in Figs. 4 and 5. The sharp features in the visible-UV derivative spectra in Fig. 4 correspond to critical points E_1 , $E_1 + \Delta_1$, E'_0 , and E_2 . A fit using Eq. (1) gives all the critical point parameters associated with these four critical points. The main derivative feature in the infrared range in Fig. 5(a) and 5(b) corresponds to E_0 critical point with its associated parameters given by Eq. (2). The E_0 feature can be clearly seen in the first-order derivatives, and nearly identical parameters are obtained with and without smoothing operation. On the contrary, E_0 feature in the second-order derivatives appears to be overwhelmed by noise in the experimental data. We needed about 21 smoothing coefficients corresponding to a polynomial of order 5 to obtain good signal to noise ratio for second-order derivatives. E_0 obtained from first-order derivatives lies within the experimental error (3 meV) of the energy value obtained using second-order derivatives.

IV. RESULTS AND DISCUSSION

A. E_1 and $E_1 + \Delta_1$ transitions

The critical point parameters obtained for E_1 and $E_1 + \Delta_1$ for InSb film and InSb substrate samples are

FIG. 4. Numerical second derivatives of (a) real and (b) imaginary parts of the visible-UV dielectric function of an InSb film. Experimental data are plotted in circles, and the model data are plotted using solid lines.

summarized in Table I, along with reference values from the literature. The amplitudes, broadenings, energy transitions, and phase angles are in good agreement with the bulk values suggesting that the film is of good crystalline quality. The energy transitions appear to show a blue shift of 5 meV with respect to the bulk values. HRXRD indicates presence of very small compressive strain in the epitaxial film. We estimated the in-plane compressive strain in the film from the average of E_1 and $E_1 + \Delta_1$ using the deformation potential theory.²⁶ The energy shift for the average of E_1 and $E_1 + \Delta_1$ can be expressed as²⁷

$$\Delta E = (2/\sqrt{3})D_1^1(1 - c_{12}/c_{11})\varepsilon_{||}, \quad (3)$$

where $D_1^1 = -6.9$ (Ref. 28) is the hydrostatic deformation potential, $c_{11} = 6.67 \times 10^{11}$ dynes/cm² (Ref. 29), and $c_{12} = 3.65 \times 10^{11}$ dynes/cm² (Ref. 29) for InSb. $\Delta E = 5$ meV for

FIG. 5. Numerical first derivatives of (a) real and (b) imaginary parts of the infrared dielectric function of an InSb film. Experimental data are plotted in circles, and the model data are plotted using solid lines.

TABLE I. Critical point parameters associated with E_1 and $E_1 + \Delta_1$ transitions for the InSb film.

Parameter	InSb film	Bulk InSb	
	This work	This work	Other reports
E_1 (eV)	1.862 ± 0.002	1.857 ± 0.002	$1.872^a, 1.845^b, 1.86^c$
A_{E_1}	$4.6 \pm 0.3, 4.0^d$	$5.1 \pm 0.3, 4.0^d$	
Γ_{E_1} (eV)	0.054 ± 0.002	0.050 ± 0.002	0.05^c
Φ_{E_1} (radians)	1.39 ± 0.06	1.41 ± 0.05	1.48^c
$E_1 + \Delta_1$ (eV)	2.367 ± 0.008	2.362 ± 0.006	$2.374^a, 2.362^b, 2.36^c$
Δ_1 (eV)	0.505 ± 0.008	0.505 ± 0.006	$0.502^a, 0.517^b, 0.50^c$
$A_{E_1 + \Delta_1}$	$2.8 \pm 0.5, 2.7^d$	$3.0 \pm 0.4, 2.7^d$	
$\Gamma_{E_1 + \Delta_1}$ (eV)	0.080 ± 0.008	0.071 ± 0.006	0.072^c
$\Phi_{E_1 + \Delta_1}$ (radians)	1.5 ± 0.2	1.5 ± 0.1	1.57^c
$\frac{A_{E_1}}{A_{E_1 + \Delta_1}}$	$1.6, 1.5^d$	$1.7, 1.5^d$	1.8^c

^aExperimental values from Ref. 14.

^bExperimental values from Ref. 13.

^cExperimental values from Ref. 11.

^dTheoretical values obtained using Eqs. (4) and (5).

the InSb film with respect to bulk InSb. We obtain $\varepsilon_{||} \sim -0.1\%$, which indicates that the film is $>99\%$ relaxed as determined by HRXRD. Furthermore, spin orbit splitting parameter Δ_1 associated with the $E_1/E_1 + \Delta_1$ critical point is 505 meV, which is in good agreement with the reported values.

The amplitudes of E_1 and $E_1 + \Delta_1$ transitions denoted by A_{E_1} and $A_{E_1 + \Delta_1}$, respectively, are summarized in Table I along with their ratio $\frac{A_{E_1}}{A_{E_1 + \Delta_1}}$. We obtain $\frac{A_{E_1}}{A_{E_1 + \Delta_1}} \sim 1.6$ and 1.7 for film and bulk InSb, respectively, which is in good agreement with the experimental ratio¹¹ of ~ 1.8 reported for bulk InSb. This can be also compared with the theoretical ratio obtained from $k.p$ theory. The theoretical expressions for A_{E_1} and $A_{E_1 + \Delta_1}$ are given by¹¹

$$A_{E_1} \simeq \frac{44 \left(E_1 + \frac{\Delta_1}{3} \right)}{a_0 E_1^2}, \quad (4)$$

$$A_{E_1 + \Delta_1} \simeq \frac{44 \left(E_1 + \frac{2\Delta_1}{3} \right)}{a_0 (E_1 + \Delta_1)^2}, \quad (5)$$

where a_0 is the lattice constant of InSb. The $k.p$ theory predicts the amplitude ratio to be ~ 1.5 . This is consistent with the experimental amplitude ratios obtained for bulk InSb and InSb film.

The broadening parameters for E_1 and $E_1 + \Delta_1$ are slightly larger than the bulk values obtained in this work as well as those reported for bulk in literature. This might be attributed to the presence of threading dislocations and small compressive strain in the nearly relaxed films.^{27,30} The phase angles for E_1 and $E_1 + \Delta_1$ obtained for the film are in good agreement with the bulk values. We notice that the magnitude of phase angle for E_1 is slightly greater than that for $E_1 + \Delta_1$ for both our film and bulk, consistent with the finding for bulk InSb¹¹ at room temperature.

TABLE II. Critical point parameters associated with E'_0 and E_2 transitions for the InSb film.

Parameter	InSb film		Bulk InSb	
	This work		This work	Other reports
E'_0 (eV)	3.14 ± 0.14		3.14 ± 0.14	$3.161^a, 3.16^b$
$A_{E'_0}$	1.7 ± 1		1.6 ± 1	...
$\Gamma_{E'_0}$ (eV)	0.21 ± 0.14		0.20 ± 0.14	...
$\Phi_{E'_0}$ (radians)	-1.3 ± 1		-1.2 ± 1	...
E_2 (eV)	4.05 ± 0.01		4.03 ± 0.01	$4.034^a, 4.08^b$
A_{E_2}	8.3 ± 1.3		9.3 ± 1.4	
Γ_{E_2} (eV)	0.16 ± 0.01		0.16 ± 0.01	0.157^b
Φ_{E_2} (radians)	2.7 ± 0.2		2.7 ± 0.2	2.79^b

^aExperimental values from Ref. 32.^bExperimental values from Ref. 11.

B. E'_0 and E_2 transitions

The critical points associated with E'_0 and E_2 are summarized in Table II. E'_0 corresponds to two doublets but is modeled as a single critical point in our analysis, because we are not able to resolve the weak internal doublet structures from this room temperature data. It has been reported that these features stay well resolved in the second derivative spectra up to 200 K.¹³ Low temperature investigation of E'_0 in bulk InSb reveals all its associated features and is easier to track them up to room temperature. The E'_0 transition value obtained for InSb film is consistent with the reported values for bulk InSb. The critical parameters associated with E'_0 show much larger uncertainties compared to other critical points. This is reported to be true for even temperatures lower than room temperature.¹³

Unlike E'_0 critical point, E_2 critical point is a strong feature in the UV energy range. The energy transition value for the film is in good agreement with our bulk InSb value as well as the bulk values reported in literature. The broadenings and phase angles obtained for this critical point in InSb film agree well with the bulk values. The amplitude of the E_2 transition has not been reported but the value obtained for the film is consistent with the value we obtained for the bulk.

C. E_0 transition and absorption coefficient

The critical point parameters obtained for E_0 for InSb film and bulk InSb samples are summarized in Table III. The bandgap value from the first derivative analysis agrees well

TABLE III. Critical point parameters associated with the direct bandgap E_0 for the InSb film.

Parameter	InSb film		Bulk InSb	
	This work	Other reports	This work	Other reports
E_0 (eV)	0.175 ± 0.001	0.175^a	0.178 ± 0.001	$0.17^b, 0.18^c$
A_{E_0} (eV) ^{-1/2}	1.39 ± 0.05	...	0.77 ± 0.04	...
Γ_{E_0} (eV)	0.029 ± 0.001	...	0.010 ± 0.001	...
Φ_{E_0} (radians)	2.49 ± 0.04	...	2.62 ± 0.05	...

^aExperimental value from Ref. 33.^bExperimental value from Ref. 29.^cExperimental value from Ref. 32.

FIG. 6. Infrared absorption coefficient of an InSb film grown on GaAs (001). Absorption coefficient of bulk InSb (Ref. 31) is shown for comparison. The absorption coefficient of the film nearly matches that of bulk InSb.

with the reported values for InSb obtained from absorption and photoluminescence measurements. The energy transition and phase angle for the film are in good agreement with the values obtained for bulk. The broadening parameter for the film is nearly three times larger than its counterpart in bulk. As pointed out for $E_1/E_1 + \Delta_1$ transition, this may be related to residual strain in the film as well as the presence of threading dislocations in bulk of the film. Also, the broadening parameter associated with E_0 might be more sensitive to threading dislocations and strain in comparison to $E_1/E_1 + \Delta_1$ transition. Fig. 6 shows the absorption coefficient for the film with the absorption coefficient for bulk InSb taken from Ref. 31. There is good agreement between the two. We notice that the absorption edge for bulk appears to be sharper than the absorption edge for the film. This is consistent with the larger broadening parameter found in the film.

V. CONCLUSIONS

In conclusion, we studied the optical properties of an InSb film grown on GaAs (100) using spectroscopic ellipsometry. The complex dielectric function of InSb film is determined from 0.06 to 4.6 eV. The critical points E_1 , $E_1 + \Delta_1$, E'_0 , and E_2 are analyzed using a second derivative analysis of dielectric function. A first derivative analysis was used to obtain the critical point parameters associated with E_0 . The results indicate that the film is of optical quality similar to bulk InSb and is nearly strain-free.

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