Faculty and Staff Publication:

**Manasa Medikonda, Gangadhara R. Muthinti, Relja Vasić,
Thomas N. Adam, Alexander Reznicek,
Matthew Wormington, Girish Malladi, Yihwan Kim,
Yi-Chiau Huang, and Alain C. Diebold**

Citation for Article:

Medikonda, M., Muthinti, G. R., Vasić, R., Adam, T. N., Reznicek, A., Wormington, M., ...
Diebold, A. C. (2014). Optical properties of pseudomorphic $\text{Ge}_{1-x}\text{Sn}_x$ ($x = 0$ to 0.11) alloys
on Ge(001). *Journal of Vacuum Science & Technology B, Nanotechnology and
Microelectronics: Materials, Processing, Measurement, and Phenomena*, 32, 061805.
doi:10.1116/1.4901254

<http://avs.scitation.org/doi/full/10.1116/1.4901254>

Optical properties of pseudomorphic $\text{Ge}_{1-x}\text{Sn}_x$ ($x = 0$ to 0.11) alloys on $\text{Ge}(001)$

Manasa Medikonda, Gangadhara R. Muthinti, Relja Vasić, and Thomas N. AdamAlexander ReznicekMatthew WormingtonGirish MalladiYihwan Kim and Yi-Chiau HuangAlain C. Diebold

Citation: *Journal of Vacuum Science & Technology B, Nanotechnology and Microelectronics: Materials, Processing, Measurement, and Phenomena* **32**, 061805 (2014); doi: 10.1116/1.4901254

View online: <http://dx.doi.org/10.1116/1.4901254>

View Table of Contents: <http://avs.scitation.org/toc/jvb/32/6>

Published by the [American Vacuum Society](#)

Instruments for Advanced Science

Contact Hiden Analytical for further details:

W www.HidenAnalytical.com
E info@hiden.co.uk

CLICK TO VIEW our product catalogue

Gas Analysis

- › dynamic measurement of reaction gas streams
- › catalysis and thermal analysis
- › molecular beam studies
- › dissolved species probes
- › fermentation, environmental and ecological studies

Surface Science

- › UHV TPD
- › SIMS
- › end point detection in ion beam etch
- › elemental imaging - surface mapping

Plasma Diagnostics

- › plasma source characterization
- › etch and deposition process reaction
- › kinetic studies
- › analysis of neutral and radical species

Vacuum Analysis

- › partial pressure measurement and control of process gases
- › reactive sputter process control
- › vacuum diagnostics
- › vacuum coating process monitoring

Optical properties of pseudomorphic $\text{Ge}_{1-x}\text{Sn}_x$ ($x = 0$ to 0.11) alloys on Ge(001)

Manasa Medikonda,^{a)} Gangadhara R. Muthinti, Relja Vasić, and Thomas N. Adam
College of Nanoscale Science and Engineering, SUNY, Albany, New York 12203

Alexander Reznicek
IBM Research at Albany Nanotech, Albany, New York 12203

Matthew Wormington
Jordan Valley Semiconductors, Austin, Texas 78744

Girish Malladi
College of Nanoscale Science and Engineering, SUNY, Albany, New York 12203

Yihwan Kim and Yi-Chiau Huang
Applied Materials, Sunnyvale, California 94085

Alain C. Diebold
College of Nanoscale Science and Engineering, SUNY, Albany, New York 12203

(Received 3 June 2014; accepted 27 October 2014; published 19 November 2014)

The characterization of the optical properties of pseudomorphic $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($x = 0$ to 0.11) alloys from the IR to UV is presented. The $\text{Ge}_{1-x}\text{Sn}_x$ alloys were epitaxially grown on relaxed Ge grown on Si. Rutherford backscattering (RBS) and RBS ion channeling methods were used to confirm the Sn composition and substitutional nature of the Sn into the Ge lattice. The pseudomorphic nature of the $\text{Ge}_{1-x}\text{Sn}_x$ on Ge is confirmed using high resolution x-ray diffraction (HRXRD) and transmission electron microscopy. Although HRXRD reciprocal space maps indicated that the $\text{Ge}_{1-x}\text{Sn}_x$ was pseudomorphic to Ge, the shape of the Bragg peaks indicated that the sample surface was rough. The rough surface morphology is confirmed using atomic force microscopy. The complex dielectric function is reported in the IR, visible, and UV spectrum in the wavelength range of $0.2\text{--}5.06\text{ eV}$. The E_1 , $E_1 + \Delta_1$, E_2 , and E_0 critical points are extracted using second and third derivative line shape fitting and are compared with the elastic theory calculations of strained $\text{Ge}_{1-x}\text{Sn}_x$ ($x = 0$ to 0.11) alloys and fully relaxed $\text{Ge}_{1-x}\text{Sn}_x$ ($x = 0$ to 0.11) alloys. The E_0 critical point energies are observed to have slightly larger values than those calculated for completely relaxed $\text{Ge}_{1-x}\text{Sn}_x$ alloys due to the presence of compressive strain. © 2014 American Vacuum Society. [<http://dx.doi.org/10.1116/1.4901254>]

I. INTRODUCTION

The optical and electronic properties of germanium (Ge) based group IV materials have potential applications in novel electronic and optoelectronic devices.^{1–3} Often the viability of these applications requires process based strain engineering or alloying. Germanium's high hole mobility can be improved through strain engineering, and its small, indirect band gap can be transformed into a direct band gap material by alloying with other elements including tin (Sn). Strain free $\text{Ge}_{1-x}\text{Sn}_x$ alloys formed by substituting Sn into Ge lattice were found to transform from indirect band gap as the percentage of Sn increased.^{4,5} Ge has very small energy difference of 0.14 eV between the lowest direct band gap and indirect band gap. The addition of Sn affects the band structure of Ge, which results in the reduction of the direct band gap (Γ -valley) more than the indirect band gap (L-valley).^{1,6} The large lattice mismatch of about 14.9% between Ge and Sn and about 19.4% between Si and Sn introduces strain in the GeSn lattice when it is epitaxially grown on Si or Ge. This strain also alters the band structure.^{7,8} Strain free

and strained $\text{Ge}_{1-x}\text{Sn}_x$ alloys with a wide range of Sn composition values have been reported in the literature.^{2,8–12} Theoretical predictions of the compositional dependence of the crossover to a direct band gap for strain free $\text{Ge}_{1-x}\text{Sn}_x$ alloys range from 6.3% ($x = 0.063$) to more than 21% ($x = 0.21$).^{2,9–12} The different crossover values are due to the specific band structure calculation method and different bowing parameters used to estimate band gaps. Recent photoluminescence studies indicate that the transition to direct band gap occurs between 6% and 8% Ge for strain free alloys.^{12,13} Gupta *et al.* report that the inclusion of alloy disorder in the crystal potential and the effect of room temperature results in a theoretical prediction of 6.5% Ge for indirect to direct band gap transition for strain free alloys.¹⁴ Strain engineering of Ge through alloying with Sn has shown to have a number of applications in MOS devices including as a stressor material for mobility enhancement which increases switching speed.^{15,16} However, several challenges including the extremely low Sn solid solubility of about 1% in Ge, low surface free energy and high surface segregation of Sn need to be addressed before growth of high quality single crystal GeSn alloys is possible. Recent advances in epitaxial growth techniques have enabled fabrication of

^{a)}Electronic mail: mmedikonda@albany.edu

pseudomorphic GeSn alloys on Ge.¹⁵ Gupta *et al.* also predict that the biaxial stress in pseudomorphic Ge_{1-x}Sn_x/Ge (001) will not result in the transition to a direct band gap for $x < 0.15$, while Ragan and Atwater claim that the indirect to direct energy transition occurs near $x = 0.09$ for epitaxial GeSn alloys coherently grown on Ge (001).^{14,17} In this paper, characterization of the crystal structure, complex dielectric function, and band gap of pseudomorphic Ge_{1-x}Sn_x alloys is reported.

Optical measurement of film thickness is a crucial process monitoring technique during the fabrication of integrated circuits. Film thickness measurement for Ge_{1-x}Sn_x alloys is complicated by the changes in the complex refractive index (dielectric function) due to strain. The optical properties of fully relaxed Ge_{1-x}Sn_x alloys for Sn concentrations up to $x = 0.2$ in the range of 0.2–6 eV and for pseudomorphic Ge_{1-x}Sn_x alloys ($x < 0.2$) in the range of 1.2–4.7 eV have been previously reported.^{4,6,8,18,19} It is useful to first discuss the optical properties of Ge and Sn. The dielectric function of Ge has several sharp features that are due to direct band transitions known as critical points (CPs).²⁰ CPs occur when there is nearly constant energy separation between the valence band and the conduction band.^{20–22} The imaginary part of the dielectric function (ϵ_2) of group IV materials exhibit Van Hove singularities or critical points M_0 , M_1 , M_2 , and M_3 .^{21,23} The E_0 CP is the lowest direct band energy due to transitions at the Γ point ($\Gamma_{8v^+} - \Gamma_{7c^-}$) of Brillouin zone.²⁴ For Ge, the E_0 CP is about 0.80 eV, which is slightly above the lowest indirect gap energy of 0.66 eV ($\Gamma_{8v^+} - L_{6c^-}$) and for Sn, E_0 occurs around -0.41 eV ($\Gamma_{8v^+} - L_{7v^-}$).^{6,24} The spin orbit splitting, Δ_1 for both germanium and tin are large enough to be observed in the dielectric function. The E_1 CP arises due to the transition from highest valence band to lowest conduction band along the $\langle 1, 1, 1 \rangle$ direction.²³ E_1 CP occurs at 2.12 and 1.27 eV for Ge and Sn, respectively, with $E_1 + \Delta_1$ CPs at 2.31 and 1.77 eV.^{6,22} The spin orbit split value, which depends on the bands involved in the transition, exhibits Δ_1 value for Ge as approximately 0.19 eV and for Sn as 0.5 eV.⁶ The critical points and spin orbit splits for Ge_{1-x}Sn_x alloys can be approximated using a quadratic polynomial and composition dependent bowing parameters.⁶

Several papers reported the optical properties of strain free CVD grown GeSn alloys grown on Si.^{18,19} Recently, the optical properties of fully strained GeSn alloys on Ge grown by MBE were reported in the visible region.⁸ The authors are not aware of a report of the dielectric function of CVD grown pseudomorphic Ge_{1-x}Sn_x alloys in the IR region. In this paper, we report the optical properties of CVD grown Ge_{1-x}Sn_x ($x < 0.11$) alloys pseudomorphic to Ge from the IR to the UV region (from 0.2 to 5.06 eV). This paper also studies the stress induced shifts of the E_1 critical point using deformation potentials that vary with composition. The structure of the GeSn alloys is a function of the specific process used to fabricate the epilayers. This paper describes the results of the characterization of pseudomorphic GeSn alloys that were fabricated on 300 mm wafers using equipment and processes that are compatible with current semiconductor integrated circuit manufacturing. Since changes in processing conditions and

substrate can alter the resulting epilayer, the films were thoroughly characterized. The pseudomorphic nature of the films used in this study was confirmed using high resolution x-ray diffraction (HRXRD), and the results are also presented. This article also discusses the absence of interference fringes in ω -2 θ HRXRD scans and reciprocal space maps (RSMs), which is attributed to the surface roughness and defects present at the Si/Ge interface. This conclusion is supported by the atomic force microscopy (AFM) and transmission electron microscopy (TEM) images. In Sec. II, the elastic theory describing shift in the CP energy with the increase in Sn concentration is discussed. The experimental methods are described in Sec. III. In Sec. IV, the characterization results of the Ge_{1-x}Sn_x alloys using AFM, TEM, Rutherford backscattering (RBS), HRXRD and spectroscopic ellipsometry (SE) are presented. The RBS determination of Sn composition in the Ge_{1-x}Sn_x alloys is described along with high resolution x-ray diffraction confirmation. The quality of the crystalline material and the Sn substitution in the Ge lattice is also studied using HRXRD and RBS analysis. Detailed discussion of the absence of interference fringes in HRXRD of fully strained pseudomorphic GeSn layers is also presented. Finally, the complex dielectric functions derived from the ellipsometric measurements are also presented. Conclusions are provided in Sec. V.

II. ELASTIC THEORY MODEL FOR STRESS INDUCED CHANGES IN THE OPTICAL PROPERTIES

Prediction of the stress induced shift of CPs of biaxially strained Ge_{1-x}Sn_x alloys is presented in this section.^{6,8} Ge_{1-x}Sn_x alloys up to $x = 0.2$ were shown to no phase separation into GeSn alloy and excess Sn based on HRXRD measurements including 004 ω -2 θ rocking curves presented below.⁸

The room temperature lattice constant of unstrained, bulk Ge_{1-x}Sn_x is known to be⁶

$$a(\text{Ge}_{1-x}\text{Sn}_x) = a_0(\text{Sn})x + a_0(\text{Ge})(1-x) + \theta_{\text{GeSn}}(1-x)x, \quad (1)$$

where θ_{GeSn} indicates the bowing parameter, which contributes to the deviation from Vegard's law. Different bowing values were published for GeSn alloys. Kouvetakis *et al.*⁶ reported $\theta_{\text{GeSn}} = 0.166 \text{ \AA}$, Chibane and Ferhat²⁵ reported $\theta_{\text{GeSn}} = 0.65 \text{ \AA}$, Beeler *et al.*²⁶ extracted it to be $\theta_{\text{GeSn}} = 0.0468 \text{ \AA}$, and Gencarelli *et al.*²⁷ calculated it out to be $\theta_{\text{GeSn}} = 0.041 \text{ \AA}$. Here, we use the bowing value of 0.0468 $\text{\AA}$. It is important to note that the bowing parameter of GeSn alloys has the opposite sign to that observed for Si_{1-x}Ge_x alloys.⁶

Based on the elastic theory, the in-plane strain component $\epsilon_{\perp}$ and the perpendicular strain component $\epsilon_{\parallel}$, i.e., in the growth direction are given by^{8,20,22,28}

$$\epsilon_{\perp} = \frac{a_{\text{Ge}} - a_{\text{GeSn}}}{a_{\text{GeSn}}} \quad \text{and} \quad \epsilon_{\parallel} = \frac{2S_{12}}{S_{11} + S_{12}}\epsilon_{\perp} = -\frac{2C_{12}}{C_{11}}\epsilon_{\perp}. \quad (2)$$

In this paper, notations for the in-plane direction and the growth direction follow Zollner's review of k*p theory of strain dependence of the optical properties of Si_{1-x}Ge_x alloys. The elastic stiffness constants C_{11} , C_{12} and the elastic compliance constants S_{11} , S_{12} are related by $C_{11} - C_{12} = (S_{11} - S_{12})^{-1}$ and $C_{11} + 2C_{12} = (S_{11} + 2S_{12})^{-1}$. For Ge_{1-x}Sn_x alloys, these elastic compliance constants are estimated using Vegard's law (Table I).

The room temperature CP energies E_1 and $E_1 + \Delta_1$ of strain-free Ge_{1-x}Sn_x alloys depend on the Sn concentration and follow the quadratic polynomial expression

$$E_i(x) = E_i^{Sn} x + E_i^{Ge} (1 - x) - b_i x (1 - x), \quad (3)$$

where b_i is again the bowing parameter defining the deviation from the linear interpolation of the Ge and Sn band gaps. Experimental b_i values were reported to be around 2.1–2.8 eV.^{4,12} The spin orbit splitting, Δ_1 of Ge and Sn can be observed in ellipsometrically determined dielectric functions. Thus, the spin orbit splitting of Ge_{1-x}Sn_x alloys can also be observed.

Biaxially strained tetrahedral semiconductors have shown to exhibit E_1 and $E_1 + \Delta_1$ band gaps following the expressions:^{8,20,22,28}

$$E_1(x, \varepsilon) = E_1(x) + \frac{\Delta_1(x)}{2} + \Delta E_H - \sqrt{\Delta E_S^2 + \left(\frac{\Delta_1(x)}{2}\right)^2}, \quad (4)$$

$$(E_1 + \Delta_1)(x, \varepsilon) = (E_1 + \Delta_1)(x) - \frac{\Delta_1(x)}{2} + \Delta E_H + \sqrt{\Delta E_S^2 + \left(\frac{\Delta_1(x)}{2}\right)^2}, \quad (5)$$

where Δ_1 is the spin orbit splitting estimated using Vegard's law and composition dependent bowing parameter for Ge_{1-x}Sn_x alloys.⁶ The predicted stress induced shift of the $E_1 + \Delta_1$ CP is larger than that predicted for the E_1 CP. ΔE_H and ΔE_S are the hydrostatic and shear shifts in the CP energies for biaxially strained Ge_{1-x}Sn_x alloys given by hydrostatic and shear strain components e_H and e_S , respectively. D_1^1 and D_3^3 are the hydrostatic and shear deformation potentials^{8,28}

$$\Delta E_H = \frac{2}{\sqrt{3}} D_1^1 \left(1 - \frac{C_{12}}{C_{11}}\right) \varepsilon_{\perp} = \sqrt{3} D_1^1 e_H, \quad (6)$$

$$e_H = \frac{\varepsilon_{\parallel} + 2\varepsilon_{\perp}}{3},$$

TABLE I. Elastic constants, phonon deformation potentials, and lattice constants for germanium and α -tin at room temperature (Refs. 7, 8, 24, and 29).

	C_{11} (10^{12} dyn cm ⁻²)	C_{12} (10^{12} dyn cm ⁻²)	D_1^1 (eV)	D_3^3 (eV)	a (Å)
Ge	1.289	0.483	-8.6	5.9	5.6579
Sn	0.690	0.293	—	—	6.4892
Ge _{0.80} Sn _{0.20}	1.1692	0.445	-5.4	3.8	5.8316

$$\Delta E_S = -\sqrt{\frac{2}{3}} D_3^3 \left(1 + 2 \frac{C_{12}}{C_{11}}\right) \varepsilon_{\perp} = \sqrt{6} D_3^3 e_S, \quad (7)$$

$$e_S = \frac{\varepsilon_{\parallel} - \varepsilon_{\perp}}{3}.$$

The hydrostatic and shear deformation potentials D_1^1 and D_3^3 for Ge_{1-x}Sn_x alloys (x < 0.20) were calculated and reported as -5.4 and 3.8 eV by D'Costa *et al.* and were considered invariant of Sn composition change for concentrations less than 20%.⁸ Here, the deformation potentials between 0% and 20% Sn concentrations are determined using linear interpolation

$$D_z^z(\text{Ge}_{1-x}\text{Sn}_x) = D_z^z(\text{Ge}) + \left[\frac{D_z^z(\text{Ge}_{0.80}\text{Sn}_{0.20}) - D_z^z(\text{Ge})}{0.20} \right] * (x). \quad (8)$$

III. EXPERIMENTAL METHODS

Strained Ge_{1-x}Sn_x (x = 0.04 to 0.11) layers were grown separately on relaxed Ge buffer layers grown on double-sided polished 300 mm p-type Boron doped (1–10 Ω cm) (100) silicon wafers using chemical vapor deposition. Digermane (Ge₂H₆) and Tin tetrachloride (SnCl₄) were used as precursors for Ge and Sn. The samples were subjected to a high temperature hydrogen bake at 1040 °C to remove any contamination on the Si substrate wafer before Ge deposition. The Ge layer was then deposited followed by an *in-situ* GeSn deposition. AFM data were profiled over an area of 10 μ m $\times$ 10 μ m for surface analysis. The Si-Ge interface and the Ge-GeSn interfaces were probed using TEM cross-section analysis to detect any dislocations or defects present at the interfaces.

Jordan Valley's BEDE Metrix-L (BML) x-ray diffractometer tool was utilized for high resolution x-ray diffraction analysis on the GeSn samples. The BML tool provides an x-ray beam of a few millimeters with a 2.2 kW Cu sealed tube source and a channel-cut collimator. The tool has an angular resolution of 25" and is capable of handling 300 mm wafers.

RBS channeling angular scans of GeSn/Ge/Si multilayers were acquired using 2 MeV He⁺ ion beam. The collected charge limit was set to 1 μ C to minimize the radiation damage effects. The charge limit for random configuration was set to 4 μ C with the detector about 7° away from the incoming beam in Ge {100} direction to avoid the channeling effect of single crystal substrate. The angular yield scans were obtained in the range of -2° to +2° from the channel. The energy spectra of He⁺ ions were measured using magnetic spectrometer.

Spectroscopic ellipsometry data was acquired using J. A. Woollam RC2 in the range of 0.73–5.06 eV and IR-VASE in the range of 0.2–0.8 eV. The ellipsometric angles Ψ and Δ were collected along with depolarization data with a resolution of 0.01 eV using the dual rotation compensator RC2 and with a resolution of 0.06 eV using IR-VASE. For IR-VASE

measurements, the samples were backside roughened to obtain the reflected signal.

Atomic force microscope by Digital Instruments was used to probe the surface of $\text{Ge}_{1-x}\text{Sn}_x$ alloys. An area of $100\ \mu\text{m}^2$ is scanned to determine the surface roughness of the GeSn layer.

IV. RESULTS

A. AFM and TEM Analyses

AFM characterization of surface roughness of the $\text{Ge}_{1-x}\text{Sn}_x$ alloy samples determined that the samples were considerably rougher than the sub-0.2 nm RMS observed for pseudomorphic $\text{Si}_{1-x}\text{Ge}_x/\text{Si}$.²² A line scan and an area scan on the GeSn surface of $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($x = 0.04$) sample is shown in Fig. 1. The AFM scans displayed Z_{height} of 12.13 nm with a Z_{rms} value of 1.69 nm. AFM scans of the $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($x = 0.11$) sample shown in Fig. 2 also displayed similar results with Z_{height} of 13.82 nm and a Z_{rms} value of 2.05 nm.

TEM imaging of the Si-Ge interface of $\text{Ge}_{0.89}\text{Sn}_{0.11}/\text{Ge}/\text{Si}$ sample is shown in Fig. 3. The TEM images were acquired on one region of Si-Ge interface and one region of Ge-GeSn interface. The interface between Si and Ge clearly displays high density of misfit dislocations which also migrated to the lower layers of Ge. A thick germanium buffer layer was intentionally grown so that a defect free germanium lattice is formed at the surface of the Ge. This enabled growth of high quality, fully strained pseudomorphic GeSn layers on Ge. A TEM image of the Ge-GeSn interface in Fig. 4 displays a

FIG. 1. (Color online) AFM image of the GeSn surface of the $\text{Ge}_{0.96}\text{Sn}_{0.04}/\text{Ge}/\text{Si}$ sample showing an average roughness of 1.69 nm.

FIG. 2. (Color online) AFM image of the GeSn surface of the $\text{Ge}_{0.89}\text{Sn}_{0.11}/\text{Ge}/\text{Si}$ sample showing an average roughness of 2.05 nm.

perfect GeSn lattice grown on Ge lattice with no misfit dislocations, indicating high crystalline quality of the GeSn layer.

B. RBS Analysis

RBS in the ion channeling configuration was used to evaluate the Sn concentration and crystalline quality of the

FIG. 3. TEM image of the Si-Ge interface of the $\text{Ge}_{0.89}\text{Sn}_{0.11}/\text{Ge}/\text{Si}$ sample. Misfit dislocations can be seen at the interface.

FIG. 4. TEM image of the Ge-GeSn interface of the $\text{Ge}_{0.89}\text{Sn}_{0.11}/\text{Ge}/\text{Si}$ sample displaying a defect-free strained GeSn lattice on Ge.

pseudomorphic $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($0 \leq x \leq 0.11$) samples. RBS channeling measurements were used to detect the presence of interstitial Sn in the GeSn alloy layer.

The measured random spectrum for $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($x = 0.0, 0.4$, and 0.11) samples is shown in Fig. 5. Silicon substrate spectra edge can be seen around 0.77 MeV . Large broad peaks at 1.67 MeV correspond to the germanium atoms in the Ge and GeSn layer. The width of the peak is directly proportional to the thickness of the layer. The Ge peaks cover a broad energy range due to large number of Ge atoms. The small peaks around 1.82 MeV are the Sn spectra. Increase in intensity of the tin peak can be observed in the plot as the tin concentration increases from $x = 0.04$ to $x = 0.11$. The composition and depth profiles were obtained by simulating best fits on these random spectra. The RBS thickness and composition results for all the samples $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($0 \leq x \leq 0.11$) are shown in Table II.

In order to probe for interstitial Sn, axial channeling spectra in the $\langle 100 \rangle$ channeling direction were collected for all

FIG. 5. (Color online) RBS random spectrum for samples $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($x = 0, 0.04$, and 0.11). (Inset) Sample and beam setup for collecting backscattered ions. The detector is positioned at an angle $\phi = 7^\circ$ from the incoming beam to collect backscattered ions.

samples. The backscattered ions from host and solute atoms were collected as a function of crystal alignment, which determine the lattice quality and position of solute atoms.³⁰ The channeling effects are quite distinguishable in the angular range of $\pm 2^\circ$. Channeling angular scans in the range of -2° to $+2^\circ$ were collected separately for germanium and tin of $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($x = 0.04, 0.06, 0.08$, and 0.11) samples and are shown in Fig. 6. All the scans depict very low backscattered signal of the $\langle 100 \rangle$ channel at 0° , indicating undistorted lattice structure. The Ge and Sn angular scans for $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($x = 0.11$) sample are compared in Fig. 7(a) which shows similar signal variation of Ge and Sn with angle. The angular scans were also collected for the $\langle 110 \rangle$ channel to minimize silicon substrate signal. Figure 7(b) shows Sn angular scans of $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($x = 0.11$) sample in $\langle 100 \rangle$ and $\langle 110 \rangle$ directions. The observed channeling patterns indicate substitution of Sn solute atoms in the Ge host lattice.

The ratio of aligned to random yield of the backscattered ions results in a normalized yield ($\chi^{(lmn)}$). The normalized yields in the opted channel for specific atoms in an alloy can determine the position of those atoms in the lattice.³⁰ The

TABLE II. Measured Sn concentration, Ge and GeSn layer thickness values, E_0 , E_1 , and $E_1 + \Delta_1$ critical points of pseudomorphic $\text{Ge}_{(1-x)}\text{Sn}_x$ ($x = 0.00$ to 0.11) alloys on Ge on Si substrate.

No.	Sn fraction x		Ge thickness (nm)		GeSn thickness (nm)		Strain (HRXRD) % ± 0.02	E_0 (eV) ± 0.01	E_1 (eV) ± 0.01	$E_1 + \Delta_1$ (eV) ± 0.01
	HRXRD ± 0.005	RBS ± 0.005	SE ± 0.5	RBS ± 10	SE ± 0.05	RBS ± 5				
1	0.00	0.000	641	610	0	0	0	0.8	2.1	2.32
2	0.04	0.045	641	615	77.5	80	-0.6	0.68	2.04	2.28
3	0.05	0.055	641	600	71.9	68	-0.7	0.65	2.02	2.28
4	0.06	0.060	641	600	70.5	68	-0.9	0.63	2.02	2.25
5	0.07	0.073	641	585	62.8	70	-1.0	0.6	1.99	2.22
6	0.08	0.080	641	595	60.2	68	-1.2	0.58	1.96	2.22
7	0.09	0.090	641	590	59.2	70	-1.3	0.55	1.95	2.2
8	0.10	0.110	641	580	58.3	68	-1.5	0.53	1.95	2.18
9	0.11	0.110	641	590	60.7	68	-1.6	0.51	1.9	2.21

FIG. 6. (Color online) Channeling angular scans of (a) germanium and (b) tin for $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ ($x = 0.04, 0.06, 0.08$, and 0.11) in $\langle 100 \rangle$ direction.

normalized yields of Ge and Sn for the $\langle 100 \rangle$ channel were obtained from aligned channeling spectra yield divided by the yield of random spectra. Figure 8(a) shows the normalized yields for Ge atoms and Sn atoms in the top GeSn layer of $\text{Ge}_{0.89}\text{Sn}_{0.11}/\text{Ge}/\text{Si}$ sample in the $\langle 100 \rangle$ channel. Fraction of solute atoms ($f_{\text{ds}}^{\langle 100 \rangle}$) displaced from host lattice sites into the $\langle 100 \rangle$ channel is estimated by measuring the normalized yields from host atoms ($\chi_h^{\langle 100 \rangle}$) and from solute atoms ($\chi_s^{\langle 100 \rangle}$). The fraction is determined from the formula³⁰

$$f_{\text{ds}}^{\langle 100 \rangle} = \frac{(\chi_s^{\langle 100 \rangle} - \chi_h^{\langle 100 \rangle})}{(1 - \chi_h^{\langle 100 \rangle})}. \quad (9)$$

Figure 8(b) plots the fraction (f_{ds}) of Sn atoms displaced from Ge atoms in the $\langle 100 \rangle$ channel. The results strongly suggest that the Sn solute atoms are predominantly located in Ge substitutional lattice sites.

C. X-ray characterization

High resolution x-ray diffraction was used to characterize the strain state, composition, and qualitatively analyze the defects in the GeSn alloy samples. A reciprocal space map

FIG. 7. (Color online) (a) Comparison of channeling angular scans of Ge and Sn of $\text{Ge}_{0.89}\text{Sn}_{0.11}/\text{Ge}/\text{Si}$ sample in $\langle 100 \rangle$ direction. (b) Comparison of angular scans of Sn for the same sample in $\langle 110 \rangle$ and $\langle 100 \rangle$ directions.

FIG. 8. (Color online) (a) Normalized yields of Ge and Sn for the top GeSn layer of $\text{Ge}_{0.89}\text{Sn}_{0.11}/\text{Ge}/\text{Si}$ sample in $\langle 100 \rangle$ direction. (b) Fraction of solute Sn atoms displaced from Ge host atoms into the channel estimated from the normalized yields of Ge and Sn atoms.

of 224 planes exposes the in-plane lattice orientation, which can evaluate strain/relaxation in a material. For perfectly pseudomorphic layers, the corresponding layer peak appears at the same relative reciprocal distance as that of the substrate and the shift of the layer peak from the substrate peak depends on the amount of layer relaxation.^{31–34} A 224 RSM of the $\text{Ge}_{1-x}\text{Sn}_x$ ($x = 0.11$)/ Ge/Si sample is shown in Fig. 9, indicating all the three peaks—Si substrate, Ge buffer layer, and GeSn epilayer peaks. The Q_x values in the plot clearly indicate a completely relaxed germanium layer on silicon and a strained GeSn layer with respect to germanium layer. Since the GeSn lattice is strained in the direction parallel to surface, lattice expands in the perpendicular direction, i.e., along the growth direction. Figure 10 plots Ge and GeSn

FIG. 9. (Color online) Reciprocal space map of 224 planes of $\text{Ge}_{0.89}\text{Sn}_{0.11}$ sample displaying a completely relaxed Ge layer and a pseudomorphic GeSn layer. The solid line represents the relaxation line for Ge film from Si substrate.

FIG. 10. (Color online) Reciprocal space map of (224) planes of $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ samples ($x = 0.00$ to 0.11) depicting a fully strained GeSn layer in all the samples.

peaks with Sn concentration varying from 0% to 11%. There is a clear shift of GeSn peaks away from the Ge peak in the Q_z direction as the Sn concentration increases due to the increase of GeSn lattice along the growth direction. Figure 11 depicts an 004 RSM for Sn concentrations of 4% and

FIG. 11. (Color online) Reciprocal space map of (004) planes of $\text{Ge}_{1-x}\text{Sn}_x/\text{Ge}/\text{Si}$ samples for tin concentrations of $x = 0.04$ and $x = 0.11$.

11%. 004 RSMs quantify the layer tilts present on the substrate by observing the 004 Bragg diffraction angles from both the substrate and the layer. The layer peak positions of both Ge and GeSn in the map depict a perfect untilted crystalline lattice. Defects in a lattice structure affect the diffuse scatter intensity distribution near a reciprocal lattice point (Relp).^{31,32,35} Equal peak broadening for the substrate and layers is observed because of the strain fields that penetrated into the Si substrate due to defects in the Ge epilayer.^{31,32} Holý *et al.* developed x-ray diffraction models for the mosaic structure and rough interface defects in epitaxial layers and superlattices using semikinematical approximation and optical coherence approach.³⁵ Comparing the diffuse scatter around the Relps of Ge and GeSn of our samples, there is indeed an indication of mosaic spread in the Ge layer and surface roughness for the GeSn layer. Though the $\text{Ge}_{1-x}\text{Sn}_x$ layers ($0.4 \leq x \leq 0.11$) are pseudomorphic to the Ge layer, the absence of interference fringes is attributed to the defects and roughness present in the sample.

The 004 ω - 2θ coupled line scans for all the samples are shown in Fig. 12. For a given pseudomorphic material, calculations for concentration of the substituent and the layer thickness involve rigorous computational analysis; however, the preliminary values can be estimated from intensity of the layer peak, interference fringe separation width and angular separation of the layer peak from the substrate peak.^{32,33} Due to the absence of interference fringes, calculation of the layer thickness was difficult, but the composition has been extracted using angular difference $\delta\theta$ between the substrate and the layer peaks. By comparing the experimental

FIG. 12. (Color online) ω - 2θ coupled scans of 004 planes of Ge_{1-x}Sn_x/Ge/Si samples ($x = 0.00$ to 0.11) with silicon as the reference peak. Arrows shown in the plot trace the GeSn peak which can be seen as shifting away from the Ge peak as the tin concentration increases.

mismatch (m^*) to theoretical mismatch (m_t) values of various compositions, percentage of tin content was calculated for these alloys and is within 0.5% error of the expected values. The experimental mismatch between the substrate and the layer's interplanar spacing in the growth direction is given by³¹⁻³³

$$m^* = \frac{\delta\theta}{\tan\theta + \tan\varphi}, \quad (10)$$

where θ is the Bragg angle of the reference layer and φ is the crystal plane angle. This mismatch in the direction of growth is related to the parallel mismatch by³¹⁻³³

$$m = \frac{1-\nu}{1+\nu} m^* \quad \text{and} \quad \nu = \frac{C_{12}}{C_{11} + C_{12}}, \quad (11)$$

where ν is the Poisson ratio in the $\langle 001 \rangle$ direction, C_{11} and C_{12} are the elastic stiffness coefficients. For Ge_{1-x}Sn_x alloys, the stiffness coefficients are calculated by the linear interpolation of relaxed bulk Ge and Sn values

$$C(\text{Ge}_{1-x}\text{Sn}_x) = x C_{\text{Sn}} + (1-x) C_{\text{Ge}}. \quad (12)$$

Theoretical mismatch is estimated using layer (a_l) and substrate (a_s) lattice constants. Here, mismatch between GeSn and Ge lattice is calculated for various Sn concentrations³¹⁻³³

$$m_t = \frac{a_l - a_s}{a_s}. \quad (13)$$

The theoretical mismatch values are compared with the experimental values to determine the Sn composition.

Confirming pseudomorphic nature of the GeSn layer on Ge of all the samples using HRXRD, ellipsometric studies have been done to determine the optical properties of these alloys.

D. Ellipsometry

The experimental Ψ and Δ data and the model fits for Ge_{0.96}Sn_{0.04} sample and Ge_{0.89}Sn_{0.11} sample from the IR to the UV region at different incident beam angles is shown in Fig. 13. The complex dielectric functions of the Ge_{1-x}Sn_x alloys from the IR to the UV region are shown in Figs. 14 and 15. The Ge layer and the GeSn layers in the energy range between 0.73 and 5.06 eV are modeled using Tauc-Lorentz oscillators, which ensure Kramers-Kronig consistency between the real and imaginary parts of the dielectric function. A Cauchy function consistent with germanium dioxide (GeO₂) has been included as a native oxide layer for all the samples. The dielectric function of Ge determined using the above procedure matches previously observed results of Ge.²² Although D'Costa *et al.* derived a constant value of deformation potentials for all Ge_{1-x}Sn_x ($x \leq 0.20$)⁸ alloys from the experimental results, the deformation potentials for CP transitions depend on composition.²² The deformation potentials used to calculate E_1 and $E_1 + \Delta_1$ critical points for the pseudomorphic, stressed alloys are estimated using linear interpolation between Ge and Ge_{0.80}Sn_{0.20} alloy. The CP energies are consistent with the theoretical calculations with slight differences due to the bowing parameters used in the calculation of lattice parameters and critical points. The imaginary part of the dielectric function shown in Fig. 14(b) clearly shows the E_1 , $E_1 + \Delta_1$, and E_2 critical point shifts of Ge_{1-x}Sn_x as the Sn concentration increases. The CP shifts of strained Ge_{1-x}Sn_x alloys differ from those of the unstrained alloys. The E_1 and $E_1 + \Delta_1$ CPs in Fig. 14(b) exhibit shift values comparable to the calculated CP energies for strained Ge_{1-x}Sn_x alloys. By comparing Eqs. (6) and (7), one can see that elastic theory predicts an increased splitting between E_1 and $E_1 + \Delta_1$ with increased strain (stress). Although stress increases with Sn concentration for pseudomorphic Ge_{1-x}Sn_x/Ge(001), the splitting of E_1 and $E_1 + \Delta_1$ does not increase as expected. Elastic theory predictions of the values of E_1 and $E_1 + \Delta_1$ were consistent with experimental results for pseudomorphic Si_{1-x}Ge_x/Si(001).

The dielectric functions of Ge_{1-x}Sn_x alloys in the IR region of 0.2–0.8 eV are shown in Fig. 15. Since there is a thick Ge buffer layer, which absorbs light, the light beam gets attenuated as it propagates through and traverses back.

FIG. 13. (Color online) Experimental data and model fits for (a) and (b) $\text{Ge}_{0.96}\text{Sn}_{0.04}$ sample from IR to UV region and for (c) and (d) $\text{Ge}_{0.89}\text{Sn}_{0.11}$ sample from IR to UV region. This data was obtained in the IR spectrum using the Woollam IR-SETM and in the visible to near-IR range using RC2TM.

The Ge layer, therefore, is modeled as a graded layer of 11 sublayers with the percentage of variation of optical constants between the Ge/Si interface and the Ge/air interface as fit parameters. This is likely due to a decrease in the number of defects with distance from the Ge-Si interface. The optical properties are determined by executing a point by point fit and later using Tauc–Lorentz oscillator models to match the experimental data. A surface roughness layer using an effective medium approximation is also included in all the sample models. A change in the E_0 CP was observed for $\text{Ge}_{1-x}\text{Sn}_x$ alloys with the variation of Sn content and is shown in Fig. 15. The direct energy gap E_0 due to transitions at the Γ point is predicted to decrease with the addition of Sn into Ge.

As mentioned above, Gupta *et al.* investigated the effects of biaxial strain on the band structure of $\text{Ge}_{1-x}\text{Sn}_x$ alloys using a nonlocal empirical pseudopotential method and predicted both the direct E_0 (Γ) and indirect band gap E_0 (L) energies when a combination of strain and small amounts of Sn are introduced into Ge.¹⁴ The lowest energy gap and the energy separation between the L and Γ conduction band minima ($E_{CL}-E_{C\Gamma}$) were calculated as a function of biaxial strain and Sn concentration.¹⁴ The experimental results reported in this paper were consistent with the predicted band gap values calculated by inserting the strain and

composition values from HRXRD measurements in model estimated by Gupta *et al.* The imaginary part of the dielectric function for selected samples is shown in 15(b). The dielectric functions obtained for the pure Ge and $\text{Ge}_{1-x}\text{Sn}_x$ layers in our samples is consistent with previously reported IR measurements for relaxed alloys and transmittance measurements on strained alloys.^{18,36,37} It is also reported that even though the anisotropic component of strain reduces the bandgap by splitting the light-hole and heavy-hole bands, which are degenerate at Brillouin zone center Γ for unstrained alloys, the compressive hydrostatic strain increases the average energy gap.³⁸ Therefore the strain induced E_0 CP shifts observed are very small and the CP energies are slightly larger than those of the relaxed $\text{Ge}_{1-x}\text{Sn}_x$ alloys.

To further analyze the CP features, double derivative of the dielectric function in the energy range of 0.73–5.06 eV is computed by differentiating a polynomial expression fit of the function. The third order and second order derivatives for $\text{Ge}_{0.94}\text{Sn}_{0.06}$ alloy indicating E_0 , E_1 , $E_1 + \Delta_1$, E_0' , and E_2 critical points are shown in Fig. 16, and a comparison between elastic theory and experiment is shown in Fig. 17. The generic standard critical-point line shapes are represented as^{22,39}

FIG. 14. (Color online) Complex dielectric function of Ge_{1-x}Sn_x alloys for x = 0.00, 0.04, 0.06, 0.08, and 0.11 from 0.73 to 5.06 eV. (a) Real part (ϵ_1) of dielectric function. (b) Imaginary part (ϵ_2) of the dielectric function.

$$\frac{d^2\epsilon}{dE^2} = \begin{cases} n(n-1)Ae^{i\phi}(E-E_j+i\Gamma)^{n-2}, & n \neq 0 \\ Ae^{i\phi}(E-E_j+i\Gamma)^{-2}, & n = 0 \end{cases}, \quad (14)$$

where A , E_j , Γ , and Φ represent the amplitude, threshold energy, broadening, and excitonic phase angle, respectively.³⁹ In the $\langle 111 \rangle$ direction, the upper spin-orbit valence bands and the lower conduction bands were known to be nearly parallel in zinc-blende semiconductors.^{40,41} The bands would exhibit either a 2D-M₀ CP structure if the bands are parallel along $\langle 111 \rangle$ or a 3D-M₁ CP structure if the bands are curved.⁴¹ The line shapes of the E_1 transitions for Ge and Sn were found to have a best fit with the 2D critical point with $n=0$.^{39,41} This is in contrast to Si where the E_1 CP is excitonic in nature.³⁹ The E_0 CPs were more likely 3D minima ($n=1/2$) in the electronic joint density of states and are determined using the third order derivatives using⁴²

$$\frac{d^3(\epsilon E^2)}{dE^3} = \sum_{j=1}^3 Ae^{i\phi}(E-E_j+i\Gamma)^{-4+n/2}. \quad (15)$$

The third order derivative was used for the E_0 critical point because the changes in the shape of the dielectric function in this energy range were subtle. The third order derivative enhanced the changes in line shape allowing determination of the energy of E_0 .

Figure 17 compares the experimentally determined E_0 , E_1 , and $E_1 + \Delta_1$ CPs with the relaxed and predicted elastic

FIG. 15. (Color online) Complex dielectric function of Ge_{1-x}Sn_x alloys for x = 0.00, 0.04, 0.07, and 0.11 from 0.2 to 0.8 eV. (a) Real part (ϵ_1) of dielectric function. (b) Imaginary part (ϵ_2) of the dielectric function.

theory shifts for strained alloys. Figure 17(a) compares the experimental E_0 CPs with the values predicted by Gupta¹⁴ and the E_0 values of relaxed Ge_{1-x}Sn_x (x = 0 to 0.11) alloys calculated using Eq. (3) with the bowing parameter determined by Kouvetakis.¹⁵ The strained alloy CP energies are higher than the CPs of relaxed alloys as predicted by Gupta. Figures 17(b) and 17(c) compare the experimental E_1 and $E_1 + \Delta_1$ CPs of strained alloys with that of the relaxed alloys calculated using the bowing parameter determined by Kouvetakis¹⁵ and the predicted elastic theory shifts for strained GeSn alloys. ET-L represents the elastic theory results calculated using linear interpolation between

FIG. 16. Third order and second order derivatives of imaginary part of the complex dielectric function Ge_{0.94}Sn_{0.06} alloy.

FIG. 17. (Color online) Comparison between experimental values of the E_0 (Γ), E_1 , and $E_1 + \Delta_1$ critical point energies for pseudomorphic, biaxially strained $\text{Ge}_{1-x}\text{Sn}_x$ alloys ($x = 0.00$ to 0.11), and elastic theory (ET) described in Sec. II. ET-C values are the elastic theory results calculated using constant deformation potentials by D'Costa *et al.* ET-L values are the elastic theory results calculated using linear interpolation between deformation potentials of Ge and Sn ($x = 0.20$).

deformation potentials of Ge and Sn ($x = 0.20$) and ET-C values are the elastic theory results calculated using constant deformation potentials determined by D'Costa *et al.*⁸ The E_1 CPs of strained GeSn alloys exhibited higher energies than the CP energies of relaxed alloys and much closer to the

predicted elastic theory results. The $E_1 + \Delta_1$ CPs of strained GeSn alloys determined in this study also exhibited higher energies than the CP energies of relaxed alloys, but deviated from the predicted elastic theory shifts for some samples. Further study on higher quality GeSn films is required to determine the cause of deviation of the $E_1 + \Delta_1$ CPs.

V. CONCLUSIONS

The optical properties of biaxially strained pseudomorphic $\text{Ge}_{1-x}\text{Sn}_x$ alloys ($x = 0.00$ to 0.11)/Ge on silicon were reported from the IR to the UV region. The strain and composition values were calculated and reported using HRXRD analysis and were confirmed using RBS random spectra. The pseudomorphic nature of the alloys was verified using HRXRD rocking curves and reciprocal space maps. The effect of surface roughness and defects on the x-ray diffraction patterns is discussed. The room temperature complex dielectric function of pseudomorphic $\text{Ge}_{1-x}\text{Sn}_x$ alloys ($x = 0.00$ to 0.11)/Ge on silicon was measured using spectroscopic ellipsometry from 0.2 to 5.06 eV using IR and visible wavelength ellipsometry. There is a clear shift in E_0 , E_1 , and $E_1 + \Delta_1$ CPs as the Sn concentration increases along with an increase in spin orbit splitting Δ_1 . The room temperature shift in the energies of E_1 and $E_1 + \Delta_1$ CP with stress closely follows elastic theory predictions and the energies of E_0 CP follow shifts predicted by Gupta *et al.*

ACKNOWLEDGMENTS

The authors acknowledge TEL Technology Center, America for their generous contribution of analysis time on the BML tool and Bakhru for the experiments in the Ion beam laboratory. The authors acknowledge the funding from the Center for Nanoscale Metrology.

- ¹M. Oehme, J. Werner, M. Gollhofer, M. Schmid, M. Kaschel, E. Kasper, and J. Schulze, *IEEE Photonics Technol. Lett.* **23**, 1751 (2011).
- ²E. Kasper, M. Kittler, M. Oehme, and T. Arguirov, *Photonics Res.* **1**, 69 (2013).
- ³G. Sun, R. A. Soref, and H. H. Cheng, *Opt. Express* **18**, 19957 (2010).
- ⁴G. He and H. A. Atwater, *Phys. Rev. Lett.* **79**, 1937 (1997).
- ⁵J. Mathews, R. T. Beeler, J. Tolle, C. Xu, R. Roucka, J. Kouvetakis, and J. Menéndez, *Appl. Phys. Lett.* **97**, 221912 (2010).
- ⁶J. Kouvetakis, J. Menéndez, and A. V. G. Chizmeshya, *Annu. Rev. Mater. Res.* **36**, 497 (2006).
- ⁷S. Adachi, *Properties of Group-IV, III-V and II-VI Semiconductors* (Wiley, New York, 2005).
- ⁸V. R. D'Costa, W. Wang, Q. Zhou, E. S. Tok, and Y.-C. Yeo, *Appl. Phys. Lett.* **104**, 022111 (2014).
- ⁹W.-J. Yin and X.-G. Gong, *Phys. Rev. B* **78**, 161203(R) (2008).
- ¹⁰D. Jenkins and J. D. Dow, *Phys. Rev. B* **36**, 7994 (1987).
- ¹¹K. Alberi, J. Blacksberg, L. D. Bell, S. Nikzad, K. M. Yu, O. D. Dubon, and W. Walukiewicz, *Phys. Rev. B* **77**, 073202 (2008).
- ¹²R. Chen, H. Lin, Y. Huo, C. Hitzman, T. I. Kamins, and J. S. Harris, *Appl. Phys. Lett.* **99**, 181125 (2011).
- ¹³G. Grzybowski, R. T. Beeler, L. Jiang, D. J. Smith, J. Kouvetakis, and J. Menéndez, *Appl. Phys. Lett.* **101**, 072105 (2012).
- ¹⁴S. Gupta, B. Magyari-Köpe, Y. Nishi, and K. C. Saraswat, *J. Appl. Phys.* **113**, 073707 (2013).
- ¹⁵J. Kouvetakis and A. V. G. Chizmeshya, *J. Mater. Chem.* **17**, 1649 (2007).
- ¹⁶S. Takeuchi *et al.*, *Solid. State. Electron.* **60**, 53 (2011).
- ¹⁷R. Ragan and H. A. Atwater, *MRS Proc.* **588**, 109 (2000).
- ¹⁸V. R. D'Costa, C. S. Cook, A. G. Birdwell, C. L. Littler, M. Canonico, S. Zollner, J. Kouvetakis, and J. Menéndez, *Phys. Rev. B* **73**, 125207 (2006).

- ¹⁹C. S. Cook, S. Zollner, M. R. Bauer, P. Aella, J. Kouvetakis, and J. Menéndez, *Thin Solid Films* **455**, 217 (2004).
- ²⁰“Optical properties and band structure of unstrained and strained $\text{Si}_{1-x}\text{Ge}_x$ and $\text{Si}_{1-x-y}\text{Ge}_x\text{C}_y$ alloys,” in *Silicon-Germanium Alloys, Growth Properties and Applications*, Optoelectronic Properties of Semiconductors and Superlattices Vol. 15, edited by S. Zollner, S. T. Pantellides, and M. O. Manasreh (Taylor and Francis, New York, 2002), p. 387.
- ²¹P. Y. Yu and M. Cardona, *Fundamentals of Semiconductors* (Springer, Berlin, 1996).
- ²²G. R. Muthinti, M. Medikonda, T. N. Adam, A. Reznicek, and A. C. Diebold, *J. Appl. Phys.* **112**, 053519 (2012).
- ²³P. Lautenschlager, P. B. Allen, and M. Cardona, *Phys. Rev. B* **33**, 5501 (1986).
- ²⁴O. Madelung, *Semiconductors: Data Handbook* (Springer, Germany, 2004).
- ²⁵Y. Chibane and M. Ferhat, *J. Appl. Phys.* **107**, 053512 (2010).
- ²⁶R. Beeler, R. Roucka, A. V. G. Chizmeshya, J. Kouvetakis, and J. Menéndez, *Phys. Rev. B* **84**, 035204 (2011).
- ²⁷F. Gencarelli *et al.*, *ECS J. Solid State Sci. Technol.* **2**, P134 (2013).
- ²⁸R. Lange, K. E. Junge, S. Zollner, S. S. Iyer, A. P. Powell, and K. Eberl, *J. Appl. Phys.* **80**, 4578 (1996).
- ²⁹R. M. Martin, *Phys. Rev. B* **1**, 4005 (1970).
- ³⁰Th. Wichert and M. L. Swanson, *J. Appl. Phys.* **66**, 3026 (1989).
- ³¹D. K. Bowen and B. K. Tanner, *X-ray Metrology in Semiconductor Manufacturing* (CRC, Taylor & Francis Group, Boca Raton, FL, 2006).
- ³²D. K. Bowen and B. K. Tanner, *High Resolution X-ray Diffractometry and Topography* (CRC, Tylor & Francis Group, Boca Raton, FL, 1998).
- ³³P. F. Fewster, *X-ray Scattering from Semiconductors* (Imperial College, London, 2000).
- ³⁴M. Medikonda, G. R. Muthinti, J. Fronheiser, V. Kamineni, M. Wormington, K. Matney, T. N. Adam, E. Karapetrova, and A. C. Diebold, *J. Vac. Sci. Technol., B* **32**, 021804 (2014).
- ³⁵V. Holý, J. Kuběna, E. Abramof, A. Pesek, and E. Koppensteiner, *J. Phys. D: Appl. Phys.* **26**, A146 (1993).
- ³⁶H. P. L. de Guevara, A. G. Rodríguez, H. Navarro-Contreras, and M. A. Vidal, *Superficies y Vacío* **17**, 10 (2004), available at <http://www.redalyc.org/pdf/942/94217403.pdf>.
- ³⁷V. R. D’Costa, Y. Fang, J. Mathews, R. Roucka, J. Tolle, J. Menéndez, and J. Kouvetakis, *Semicond. Sci. Technol.* **24**, 115006 (2009).
- ³⁸H. Lin, *Growth and Characterization of GeSn and SiGeSn Alloys for Optical Interconnects* (Stanford University, Stanford, 2012).
- ³⁹P. Lautenschlager, M. Garriga, L. Viña, and M. Cardonna, *Phys. Rev. B* **36**, 4821 (1987).
- ⁴⁰M. L. Cohen and T. K. Bergstresser, *Phys. Rev.* **141**, 789 (1966).
- ⁴¹S. M. Kelso, D. E. Aspnes, M. A. Pollack, and R. E. Nahory, *Phys. Rev. B* **26**, 6669 (1982).
- ⁴²E. Ferrieu, F. Beck, and D. Dutartre, *Solid State Commun.* **82**, 427 (1992).