

# Composition Quantification of SiGeSn Alloys through Time-of-Flight Secondary Ion Mass Spectrometry: Calibration Methodologies and Validation with Atom Probe Tomography

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**Abstract**—Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) is a powerful technique for elemental compositional analysis and depth profiling of materials. However, it encounters the problem of matrix effects that hinder its application. In this work, we introduce a pioneering ToF-SIMS calibration method tailored for  $\text{Si}_x\text{Ge}_y\text{Sn}_z$  ternary alloys.  $\text{Si}_x\text{Ge}_{1-x}$  and  $\text{Ge}_{1-z}\text{Sn}_z$  binary alloys with known compositions are used as calibration reference samples. Through a systematic SIMS quantification study of SiGe and GeSn binary alloys, we unveil a linear correlation between secondary ion intensity ratio and composition ratio for both SiGe and GeSn binary alloys, effectively mitigating the matrix effects. Extracted relative sensitivity factor (RSF) value from  $\text{Si}_x\text{Ge}_{1-x}$  ( $0.07 < x < 0.83$ ) and  $\text{Ge}_{1-z}\text{Sn}_z$  ( $0.066 < z < 0.183$ ) binary alloys are subsequently applied to those of  $\text{Si}_x\text{Ge}_y\text{Sn}_z$  ( $0.011 < x < 0.113$ ,  $0.863 < y < 0.935$  and  $0.023 < z < 0.103$ ) ternary alloys for elemental compositions quantification. These values are cross-checked by Atom Probe Tomography (APT) analysis, an indication of the great accuracy and reliability of as-developed ToF-SIMS calibration process. The proposed method and its reference sample selection strategy in this work provide a low-cost as well as simple-to-follow calibration route for SiGeSn composition analysis, thus driving the development of next-generation multifunctional SiGeSn-related semiconductor devices.

**Index Terms**—SiGeSn, Time-of-flight secondary ion mass spectrometry, Atom Probe Tomography, Matrix effect suppression.

## I. INTRODUCTION

SILICON-GERMANIUM-TIN (SiGeSn) [1], a Group IV semiconductor alloy material, is gaining attention for its tunable bandgap [2], [3], allowing for customization across near-infrared (NIR), mid-infrared (MIR), and far-infrared regions (FIR), crucial for optoelectronic [4], [5] and photonic applications [6], [7]. It demonstrates great potential to integrate

with existing mainstream silicon-based complementary metal-oxide-semiconductor (CMOS) [8] processes to enable cost-effective manufacturing and rapid commercialization, promising to advance electronic circuits [9], photonics [10], and other fields [11]. However, accurately and efficiently determining the composition of ternary alloy SiGeSn has posed a significant challenge for researchers. Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) is a powerful technique for surface analysis with the capability of providing both elemental composition analysis and in-depth information [12]. However, the technique has been still suffering from matrix effect for elemental composition analysis in the past decades [13], [14]. Since the secondary emission ionization probability of a given species depends entirely on the matrix, the secondary ion yield of that species is not only directly related to the concentration of that species in the host [15]. This phenomenon is referred to in SIMS as the matrix effect which is a real obstacle for quantitative analysis [16]. Due to the presence of matrix effects, SIMS requires highly reliable reference samples and data analysis strategies to suppress matrix effects and thus achieve high-quality quantitative capability [17], [18].

As for the progress of SIMS for Group IV materials, the studies so far have been only limited to SiGe [17]-[22]. To our knowledge, there are no systematic studies of the binary GeSn and ternary compound SiGeSn in SIMS. Two big challenges have hindered the progress of SiGeSn SIMS studies. The first is the absence of appropriate SiGeSn calibration reference samples due to the lack of techniques accurately characterizing the composition of SiGeSn. Several previous research groups have utilized X-ray diffraction (XRD) [17] or Rutherford backscattering spectrometry (RBS) [18] techniques to produce SiGe reference samples for SIMS calibration. However, XRD often provides inadequate data due to its reliance on extracting

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composition information from the linear relationship between the lattice parameters of two distinct components, thereby complicating the precise identification of the specific composition for the ternary compounds like SiGeSn [23]. Moreover, strain relaxation occurring during the growth of SiGeSn alloy introduces additional complexities [11]. In terms of techniques, a limitation of RBS is that SiGeSn is often grown on Si substrate since the purpose is Si-compatible optoelectronics. In this case, the Si signals tend to merge into that of the Si substrate, making it especially difficult to measure SiGeSn with Si composition accurately [24]. Also, RBS analysis is typically limited to probing material compositions within the top few micrometers, resulting in restricted depth resolution [25], which is not ideal for studying SiGeSn thin film compositions. Atom Probe Tomography (APT) is a recently emergent and powerful analytical technique for characterizing the composition and three-dimensional (3D) structure of materials at the atomic scale [26]. APT has expanded from its traditional use of analyzing metallic materials to alloy semiconductors such as GeSn and SiGeSn [27]-[30], providing an excellent technique for SiGeSn compositional characterization. While APT has become widely used to characterize composition in three dimensions, it has been reported that measurement conditions can impact the measured composition in materials like SiGe [31], SrTiO [32], AlGaN [33] and transition metals alloys [34]. Therefore, APT quantification requires precise control of experimental conditions, advanced data reconstruction and analysis techniques to obtain accurate compositional results. Despite these challenges, APT is considered a reliable technique for quantifying SiGeSn composition [29]. The APT data itself can serve as a good reference for calibrating or cross-checking SIMS results, which raises the hope of establishing SiGeSn standard to address the first challenge. However, APT measurements are usually time-consuming and not cost-effective since both focused ion beam (FIB) and low temperature (LT) techniques must be used during sample preparation and/or measurement [26]. Therefore, SIMS techniques and reliable calibration processes are still needed to achieve widespread use and time and cost efficiency. The second challenge is the difficulty of both calibrating and standardizing of the SIMS analysis protocols for SiGeSn ternary alloys due to the well-known matrix effects [35]. Previous SiGe research has found that the MCs method [36], where M is the element of interest combined with one or two Cs atoms, can be used to suppress the matrix effect, facilitating the SiGe calibration process. For example, D. Marseilhan et al. [37] found a good linear correlation between the ratios of ToF-SIMS positive secondary ion intensities  $\text{GeCs}_n^+$  and  $\text{SiCs}_n^+$  (where  $n = 1$  and  $2$ ), and the compositional ratio of Ge and Si elements under  $\text{Cs}^+$  sputtering beam. M.N. Drozdov et al. [17] reported that the ratios of ToF-SIMS positive secondary ion intensities  $\text{GeCs}_2^+$  and  $\text{SiCs}_2^+$  and negative secondary ion  $^{74}\text{Ge}^-$  and  $^{30}\text{Si}^-$ , both have a good linear relationship with the compositional ratio of Ge and Si elements under  $\text{Cs}^+$  sputtering beam. However, it is not known whether this approach can be

extended to either GeSn binary or SiGeSn ternary alloys. It is also unknown whether there are other unpredictable effects that make the matrix effect in GeSn and SiGeSn alloys more pronounced than in SiGe alloys. Considering the above-mentioned difficulties and challenges in quantifying the composition of SiGeSn alloys and the extremely minimal SiGeSn SIMS references, it is necessary to conduct a systematic compositional study of SiGeSn SIMS in order to solve this dilemma. Due to the availability of both ToF-SIMS and APT to our team, we could develop a feasible SIMS calibration protocols method to help SiGeSn further development.

In response to these two challenges, we propose a pioneering ToF-SIMS calibration methodology rooted in a systematic examination of SiGe and GeSn binary alloys. The secondary ion emission of  $^{28}\text{Si}^-$ ,  $^{74}\text{Ge}^-$  and  $^{120}\text{Sn}^-$  in SiGe and GeSn layers were investigated under Bi primary gun in a negative analysis mode. Combining with XRD analysis, a good linear correlation between the secondary ion intensity ratio  $^{28}\text{Si}^-/^{74}\text{Ge}^-$  and the Si/Ge composition ratio can be found in the  $\text{Si}_x\text{Ge}_{1-x}$  alloy in a large range from 0.07 to 0.83. The same linear correlation is also observed in the results for  $\text{Ge}_{1-z}\text{Sn}_z$  alloy with  $z$  ranging from 0.066 to 0.183. An attempt is made to utilize XRD calibrated SiGe as well as GeSn samples as the reference to determine the relative sensitivity factor (*RSF*) with the ratio unit between the secondary ion intensity ratio and the composition ratio. The *RSFs* are then used to convert the secondary ion intensity ratios into Si/Ge/Sn composition ratios to determine SiGeSn composition. The accuracy of the proposed calibration method is cross-checked by the complementary technique-APT. The data consistency between APT and TOF-SIMS results shows that the proposed method can effectively suppress matrix effects, which are routinely encountered in conventional ToF-SIMS analysis and thus hinder the quantification of materials. Our findings provide a robust framework for precise compositional analysis of SiGeSn alloys, which is essential for material optimization and quality control in the next-generation Group IV semiconductor device development.

## II. EXPERIMENTAL

### A. Growth condition for SiGe, GeSn and SiGeSn alloys

A Veeco/EPI Model 620 Solid State Source MBE system was employed to grow SiGe and GeSn materials with a base pressure of  $1 \times 10^{-10}$  Torr. SiGe samples (samples #1-3) with Si or Ge caps were grown using electron beam evaporation of Si and thermal evaporation of Ge. For GeSn samples (samples #4-8), both Ge and Sn beams were formed by thermal evaporation. SiGeSn samples were grown using reduced-pressure chemical vapor deposition (RPCVD) reactor and detailed growth condition can be found in Reference [10], [38]. All SiGeSn samples (samples #9-16) used in this work were single-layer SiGeSn grown on the Ge virtual substrate based on the Si substrate. All the samples reported in this work were not intentionally doped to avoid any unknown effects to ion yielding. Since all SiGe, GeSn and SiGeSn are high quality single crystals, impurity elements such as oxygen, fluorine and carbon in the epitaxial layer are minimal. The Si and Sn

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compositions of the three SiGe and six GeSn samples were determined by XRD measurements (see Table I and II for details). More detailed information about these SiGe and GeSn samples can be found in previous work [39], [40]. These samples are used and considered as SiGeSn calibration reference samples.

### B. ToF-SIMS measurement setup

ToF-SIMS analyses were conducted using a ToF SIMS 5 (ION TOF, Inc. Chestnut Ridge, NY) instrument equipped with a Bi liquid metal ion gun, Cs<sup>+</sup> sputtering gun and electron flood gun for charge compensation. Both the Bi and Cs ion columns are oriented at 45° with respect to the sample surface normal. The analysis chamber pressure during the measurement is maintained below  $5.0 \times 10^{-9}$  torr to avoid contamination of the surfaces being analyzed. In addition, the ToF-SIMS data from the top surface of about 20 nm were removed to eliminate surface contamination effects on the depth profile. For the depth profiles for SiGe (samples #1-3), GeSn (samples #4-8) and SiGeSn (samples #9-16) acquired in this study, 1 keV Cs<sup>+</sup> with current ranging 54.7 nA to 56.5 nA was used to create a 200  $\mu\text{m}$  by 200  $\mu\text{m}$  area, and the central 50  $\mu\text{m}$  by 50  $\mu\text{m}$  area was analyzed using 0.42 to 0.46 pA Bi<sub>3</sub><sup>+</sup> primary ion beam. The negative secondary ion mass spectra were calibrated using C<sup>-</sup>, CH<sup>-</sup> and CH<sub>2</sub><sup>-</sup>. In this work, a series of optimization experiments were conducted to optimize the following ToF-SIMS parameters, majorly involving elemental isotope selection, ionization efficiency comparison, and sputtering parameters, the details of which can be found in Supplementary Material Section 1.

### C. APT measurement setup

APT data of SiGeSn samples #9~12 was measured by Cameca LEAP 6000 using a 260 nm wavelength pulsed laser with 50 pJ laser pulse energy and 138 kHz pulse frequency at a base temperature of 30 K. APT data of SiGeSn samples #13~16 was obtained using Cameca LEAP 4000 using a 355 nm wavelength pulsed laser, with a laser pulse energy of 20 pJ, pulse frequency of 125 kHz at a base temperature of 30 K. Detailed information on APT data collection and statistical analysis can be found in Reference [27]. Composition results from the APT will be elaborated in the later section.

## III. RESULTS AND DISCUSSION

### A. ToF-SIMS analysis for SiGe and GeSn binary alloys

TABLE I

MATERIAL CHARACTERIZATION OF THE Si<sub>x</sub>Ge<sub>1-x</sub> SAMPLES USED IN THIS WORK

| Sample#    | Si $x_{\text{XRD}}$ | $T_{\text{cap}}/T_{\text{SiGe}}$ (nm) | Substrate |
|------------|---------------------|---------------------------------------|-----------|
| 1 (top)    | 0.070               | 32 (Ge)/380                           | Ge        |
| 1 (bottom) | 0.091               |                                       |           |
| 2 (top)    | 0.216               | 20 (Ge)/200                           | Ge        |
| 2 (bottom) | 0.236               |                                       |           |

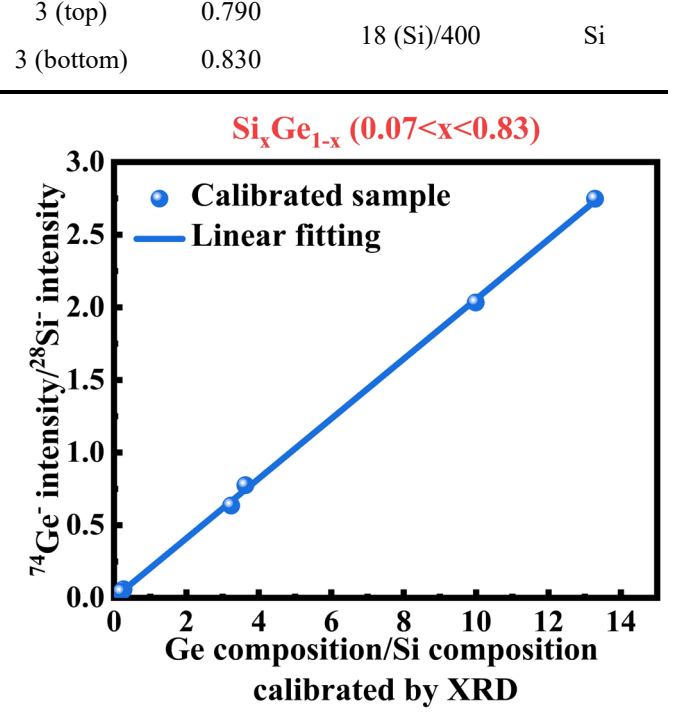


Fig. 1. The secondary ion intensity ratio  $I_{\text{Ge}}/I_{\text{Si}}$  of samples 1-3 (6 data points) versus the composition ratio calibrated by XRD and a linear fit of the calibration dependence obtained from samples 1-3.

Three Si<sub>x</sub>Ge<sub>1-x</sub> samples ( $0.07 < x < 0.83$ ) have been analyzed. Detailed layer thicknesses and compositions are shown in Table I.  $x_{\text{XRD}}$  is the Si composition of each sample as measured by XRD.  $T_{\text{cap}}$  and  $T_{\text{SiGe}}$  are the thickness of Si or Ge cap and SiGe layer. All three SiGe layers are compositional gradient layers with a Si or Ge cap layer on top of SiGe layer. The most widely used ToF-SIMS quantitative analysis technique [17] depends on finding the relative sensitivity factor ( $RSF$ ) for a given element composition. According to previous SiGe SIMS studies [36], [37], there are likely to be linear correlations between the secondary ion intensity ratio of Si related signal intensity and Ge related signal intensity and the Si/Ge composition ratio. An attempt is made to extract the  $RSF$  values using the following Eq. 1:

$$RSF * \frac{I_{\text{Ge}}}{I_{\text{Si}}} = \frac{C_{\text{Ge}}}{C_{\text{Si}}} \quad (1)$$

where  $I_{\text{Si}}$  and  $I_{\text{Ge}}$  are respectively the  $^{28}\text{Si}^-$  and  $^{74}\text{Ge}^-$  intensity values from the SiGe layer obtained by IONTOF ToF-SIMS 5 machine,  $C_{\text{Si}}$  and  $C_{\text{Ge}}$  are the Si and Ge compositions of SiGe layer calibrated by XRD. It should be noted that we used the Si and Ge compositions at the top and bottom of the SiGe layer from the three SiGe samples as data points, resulting in a total of 6 data points for the three SiGe samples. The negative ion intensity ratios of  $I_{\text{Ge}}/I_{\text{Si}}$  versus XRD-calibrated composition ratios of  $C_{\text{Ge}}/C_{\text{Si}}$  for sample #1-3 (6 data points) and the results of the  $RSF$  linear fit are shown in Fig. 1. We find a good linear correlation between the ratio of the intensity count ratios of  $^{74}\text{Ge}^-/^{28}\text{Si}^-$  and the Ge/Si composition ratio determined by XRD results. The linear regression gives a good fit with a correlation coefficient  $R^2$  value of 0.9996. The extracted  $RSF$  value for SiGe is 4.8596. The above results show the negative ion intensity ratio  $I_{\text{Ge}}/I_{\text{Si}}$  can be used to determine the SiGe alloy composition, effectively suppressing

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matrix effects. This provides a simpler method for the composition quantification of SiGe alloys. Since the Si composition ( $0.07 < x < 0.83$ ) of the  $\text{Si}_x\text{Ge}_{1-x}$  samples covers almost the entire range of  $0 \sim 1$ , this indicates that the method is applicable to the entire SiGe alloy system.

TABLE II  
MATERIAL CHARACTERIZATION OF THE  $\text{Ge}_{1-z}\text{Sn}_z$  SAMPLES  
USED IN THIS WORK

| Sample# | Sn $z_{\text{XRD}}$ | $T_{\text{GeSn}}$ (nm) | Substrate |
|---------|---------------------|------------------------|-----------|
| 4       | 0.066               | 195                    | Ge        |
| 5       | 0.077               | 320                    | Ge        |
| 6       | 0.096               | 120                    | Ge        |
| 7       | 0.156               | 40                     | Ge        |
| 8       | 0.183               | 100                    | Ge        |

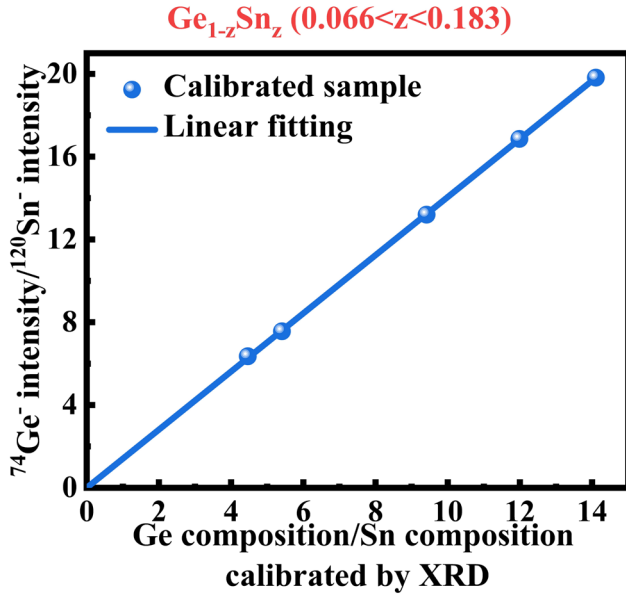


Fig. 2. The secondary ion intensity ratio  $I_{\text{Ge}}/I_{\text{Sn}}$  of samples #4-8 (5 data points) versus the composition ratio calibrated by XRD and a linear fit of the calibration dependence obtained from samples #4-8.

The same methodology as the SiGe ToF-SIMS calibration process is also applied to GeSn samples to check the matrix effect. Here, all five GeSn samples are grown without any cap layer on top and the uniformity is pretty good in terms of both composition and thickness. It should also be mentioned that sample #5 and 6 have been measured by Eurofins EAG Laboratories and the EAG result are in good agreement with the XRD results shown here. Now we are changing  $RSF$  extraction Eq. 1 for SiGe to accommodate GeSn alloys, as shown in following Eq. 2:

$$RSF * \frac{I_{\text{Ge}}}{I_{\text{Sn}}} = \frac{C_{\text{Ge}}}{C_{\text{Sn}}} \quad (2)$$

where  $I_{\text{Ge}}$  and  $I_{\text{Sn}}$  are the  $^{74}\text{Ge}^-$  and  $^{120}\text{Sn}^-$  intensity values from the GeSn layer obtained by IONTOF ToF-SIMS 5 machine,  $C_{\text{Ge}}$  and  $C_{\text{Sn}}$  are the Ge and Sn compositions calibrated by XRD. The

negative secondary ion intensity ratios  $I_{\text{Ge}}/I_{\text{Sn}}$  versus XRD-calibrated composition ratios for  $C_{\text{Ge}}/C_{\text{Sn}}$  samples #4-8 (5 data points) and the results of the  $RSF$  linear fit are shown in Fig. 2. The extracted  $RSF$  value is 0.7133. The linear regression is well fitted with a correlation coefficient  $R^2$  value of 0.9999. Very similarly, the method of obtaining composition ratios from negative ion intensity ratios is also applicable to GeSn alloys. However, it should be noted that the range of Sn composition  $z$  of  $\text{Ge}_{1-z}\text{Sn}_z$  samples is only from 0.066 to 0.183, which is a very narrow range. In the future, it is still necessary to further investigate our method feasibility to the scenarios when the Sn composition  $z$  is greater than 0.2 when those kinds of samples are available.

#### B. ToF-SIMS calibration methods and APT results for SiGeSn composition

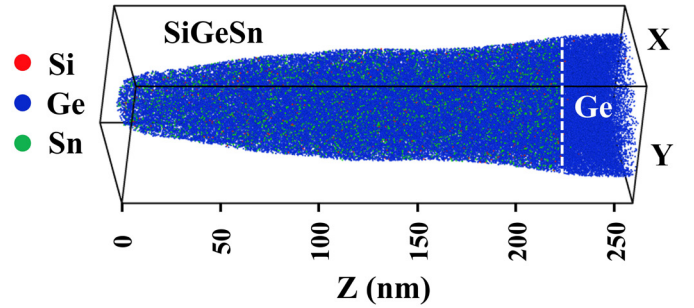


Fig. 3. Reconstructed APT data of SiGeSn sample #12. Si atoms: red dots. Ge atoms: blue dots. Sn atoms: green dots.

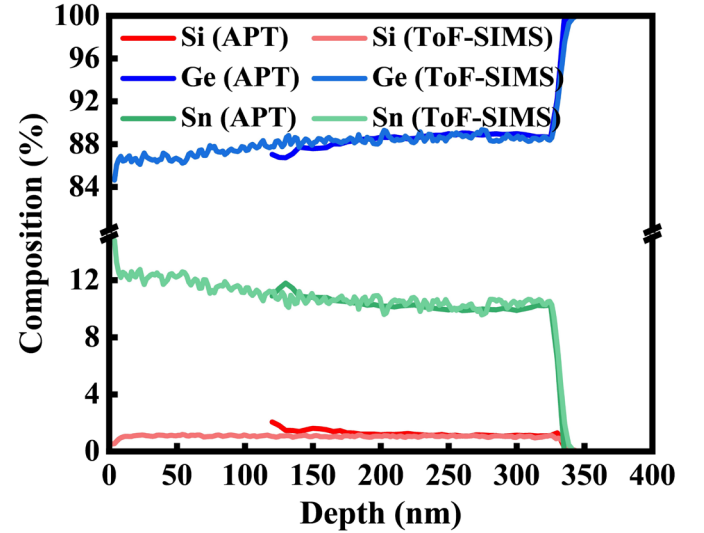


Fig. 4. Si, Ge and Sn composition depth profile from APT and ToF-SIMS in SiGeSn sample #12.

Eight SiGeSn samples grown by CVD are investigated in this work. All eight SiGeSn samples were grown on the Ge buffer layer on top of the Si substrate. Instead of using SiGeSn samples with known composition of each element directly as reference calibration samples in the conventional method, this study uses GeSn and SiGe samples as reference to calibrate the SiGeSn composition of each element. The SiGeSn samples of unknown composition were transferred into the ToF-SIMS chamber together with the SiGe and GeSn calibration samples in the

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same sample holder. First, two  $RSF$  values were determined from SiGe and GeSn samples, as shown in Fig. 1 and 2. Then, the SiGeSn samples were measured to obtain the  $^{28}\text{Si}^-$ ,  $^{74}\text{Ge}^-$ , and  $^{120}\text{Sn}^-$  intensities. The composition ratios  $C_{\text{Ge}}/C_{\text{Si}}$  and  $C_{\text{Ge}}/C_{\text{Sn}}$  will come from the negative secondary ion intensity ratios of  $I_{\text{Ge}}/I_{\text{Si}}$  and  $I_{\text{Ge}}/I_{\text{Sn}}$  based on the two extracted  $RSFs$  in the previous step. Combining all these results, it is concluded that the composition ratios of  $C_{\text{Si}}/C_{\text{Ge}}/C_{\text{Sn}}$ , as well as the final compositions of each element, can be derived. In order to verify the accuracy of our proposed ToF-SIMS calibration method for SiGeSn alloy compositions, all these SiGeSn eight samples were analyzed for elemental composition using the APT technique. Since the APT experimental setup may have some effect on the quantification of SiGeSn composition, we investigated the effect of various measurement conditions on the quantification of SiGeSn composition, as shown in Supplementary Material Section 2. The results show that the effect of measurement conditions on SiGeSn composition is very minor. The APT data can serve as a cross-check reference for evaluating and/or calibrating ToF-SIMS results. Fig. 3 is an example of an APT 3D reconstruction mapping from SiGeSn Sample #12. As shown in Fig. 3, SiGeSn Sample #12 is a SiGeSn thin film grown on a Ge buffer layer. An overlapping plot of the depth profiles of SiGeSn Sample #12 from both ToF-SIMS results and APT results is shown in Fig. 4. It can be seen that the plots for the Si, Ge and Sn compositions overlap very well. The depth profile for ToF-SIMS starts at the sample surface, however, this is not the case for APT, as FIB requires etching to form the needle-like sample shape. Therefore, a slight removal of the sample surface is unavoidable for APT technique. In order to better align the profiles from ToF-SIMS and APT, only the regions with the same distance from the SiGeSn/Ge buffer interfaces are compared.

TABLE III  
SI, GE AND SN COMPOSITIONS IN  $\text{Si}_x\text{Ge}_y\text{Sn}_z$  FILMS AS  
MEASURED WITH TOF-SIMS AND APT

| Sample#   | Si (x) | Ge (y) | Sn (z) |
|-----------|--------|--------|--------|
| 9 (SIMS)  | 0.011  | 0.932  | 0.057  |
| 9 (APT)   | 0.012  | 0.934  | 0.054  |
| 10 (SIMS) | 0.066  | 0.863  | 0.071  |
| 10 (APT)  | 0.072  | 0.860  | 0.068  |
| 11 (SIMS) | 0.034  | 0.883  | 0.083  |
| 11 (APT)  | 0.037  | 0.883  | 0.080  |
| 12 (SIMS) | 0.011  | 0.886  | 0.103  |
| 12 (APT)  | 0.012  | 0.886  | 0.102  |
| 13 (SIMS) | 0.113  | 0.864  | 0.023  |
| 13 (APT)  | 0.113  | 0.862  | 0.025  |
| 14 (SIMS) | 0.038  | 0.930  | 0.032  |
| 14 (APT)  | 0.041  | 0.927  | 0.032  |
| 15 (SIMS) | 0.024  | 0.935  | 0.041  |

|           |       |       |       |
|-----------|-------|-------|-------|
| 15 (APT)  | 0.024 | 0.936 | 0.040 |
| 16 (SIMS) | 0.075 | 0.870 | 0.055 |
| 16 (APT)  | 0.080 | 0.868 | 0.053 |

We summarize the APT and ToF-SIMS data for all eight SiGeSn samples in Table III. As can be seen, the Si and Sn compositions measured by ToF-SIMS are in good agreement with those obtained from APT, indicating the feasibility of our  $RSF$  extraction method and the calibration reference samples selection strategy. This SIMS quantification route makes the ToF-SIMS become a convenient and economical composition characterization tool for SiGeSn composition study. In addition, this calibration process can be easily extended to multilayer structures such as SiGeSn/GeSn quantum wells as well as SiGeSn with compositional grading schemes. In the future, the dopant and impurity concentrations in SiGeSn matrix will be focused based on this work. However, there are still some limitations and uncertainties in this work due to the limited composition ranges for the analyzed samples. More studies should be performed on binary of GeSn with high Sn compositions and ternary samples with high Sn and Si compositions although it is very hard to make such ones with existing technology due to the solubility limit of Sn in GeSn/SiGeSn. Further, the underlying mechanisms that suppressed the matrix effects in the as-grown SiGeSn ternary compounds are still not clear, and more theoretical studies are needed.

#### IV. CONCLUSION

In this work, ToF-SIMS analysis is applied to investigate the elemental composition of SiGe and GeSn binary alloys and SiGeSn ternary alloys. A linear correlation between the ratio of ToF-SIMS secondary ion intensities and the compositions ratio has been observed for both SiGe and GeSn. A series of SiGe and GeSn samples with the composition validated by XRD techniques are used as a reference for SiGeSn ToF-SIMS compositional studies, instead of the conventional way of using SiGeSn samples with known composition directly. Based on the  $RSFs$  value built from the SiGe and GeSn calibration reference samples, the secondary ion intensity ratios can be converted into Si/Ge/Sn composition ratios to determine the elemental compositions for SiGeSn. The introduction of the APT technique directly validates that the SiGeSn ToF-SIMS calibration method proposed in this work is feasible and reliable. The ability to characterize the elemental composition presented in this work is critical for material optimization and quality control of SiGeSn, as well as for driving the development of the next-generation Group IV-related semiconductor devices with enhanced performance and functionality.

#### APPENDIX

Appendixes, if needed, appear before the acknowledgment.

#### ACKNOWLEDGMENT

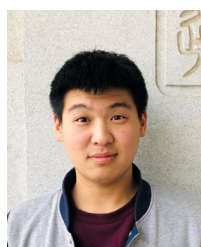
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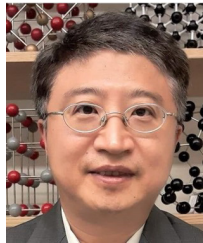


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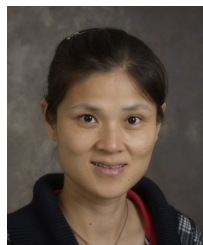
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