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Modulation of light absorption in flexible GeSn metal-semiconductor-metal photodetectors by mechanical bending

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Abstract

We have demonstrated flexible GeSn metal-semiconductor-metal (MSM) photodetectors (PDs) by exploring the effect of mechanical strain on its optoelectronic properties. The PDs were fabricated from transfer-printed GeSn nanomembranes on polyethylene terephthalate (PET) substrates. Strain was introduced into the GeSn PDs under bend-down (uniaxial tensile strain) and bend-up (uniaxial compressive strain) conditions and their values were measured by Raman spectroscopy. Applied strain can affect band-structure of GeSn alloys, leading to a modulation of electrical and optical characteristics of the PDs. Accordingly, dark current characteristics show an increase from 8.1 to 10.3 μA in the bend-down condition and a decrease to 7.2 μA in the bend-up condition, respectively. Optical responsivity at the wavelength of 2 μm was increased by 151% under bend-down condition while it decreases by 35% under bend-up condition. Theoretical study was carried out to support that the responsivity enhancement is attributed to the change in absorption coefficient of strained GeSn. These results offer a new pathway to modulate the optical properties of GeSn for flexible applications.

Introduction

Flexible optoelectronic devices have been intensively studied as a promising candidate towards various flexible and wearable applications such as flexible display¹ and low aberration optical imaging.² Among various flexible materials, semiconductor nanomembranes (NMs) are especially attractive in high performance flexible photonic applications because of the great advantages including excellent material properties (e.g., fast carrier mobility and high absorption capability) and strong bendability. Well-known examples of such semiconductor NMs are group IV (e.g., Si and Ge),^{3,4} III-V (e.g., GaAs),⁵ and wide bandgap (e.g., GaN and 4H-SiC) materials.⁶ Group IV NMs have been widely explored as active layers for enhanced light absorption and emission properties in flexible Si photonics due to the mature complementary metal-oxide-semiconductor (CMOS) process technology. Seo *et al.* have reported high performance flexible Si NM phototransistors with a back gate configuration, indicating that high absorption can be achieved from thin (i.e., 270 nm) Si NMs.⁷ Jose *et al.* have also suggested that strained Ge NMs can be an excellent material platform toward efficient group IV light emitters.⁸

Recently, germanium-tin (GeSn) alloy, another promising group-IV semiconductor, attracts much attention as one of the best candidates towards various applications in Si photonics.⁹⁻¹³ The addition of Sn into Ge allows for reduction of the bandgap energies in both Γ and L valleys and thus the efficient transition from indirect to direct band-structure via a reduction of bandgap energy difference between the Γ and L valleys ($\Delta E_{\Gamma L}$).¹⁴ High quality GeSn alloys with a Sn composition of 10 ~ 25% can be grown on Si substrates with Ge buffer layers by molecular beam epitaxy (MBE) and chemical vapor deposition (CVD).¹⁵⁻¹⁷ GeSn photodetectors (PDs) achieved a light absorption cut-off wavelength beyond 2 μm , improving light absorption in near-infrared (NIR) region while extending its applications toward mid-IR region.¹⁸ Enhanced light emission at room temperature was also observed from a GeSn vertical-cavity surface emitter grown on a silicon on insulator (SOI) substrate.¹⁹

Strain plays an important role in modulating band-structure of semiconductors, leading to enhanced performances of photonic devices. Theoretical and experimental studies have demonstrated that absorption wavelength can be further extended to 4 μm by applying tensile strain via Si_3N_4 stressors in GeSn PDs.^{20,21} Although the effect of the strain on band structure of GeSn and resultant modulation have been well analyzed, study on the effect of mechanical bending on GeSn has not been demonstrated at device level yet due to the lack of device quality flexible GeSn materials. Jin *et al.* reported the large area polycrystalline GeSn thin films on highly flexible Al foils using a MBE system.²² However, degradations on surface roughness and crystalline quality were observed due to severe Sn segregation. Flexible amorphous GeSn PD was demonstrated by Yasar *et al.* in 2018.²³ The authors formed amorphous GeSn thin films on flexible substrates by thermal evaporation of GeSn alloys. However, the devices suffered from high dark current and low light absorption due to poor crystalline quality of GeSn. In addition, experimental evidence for light absorption at NIR range was not provided.

In this paper, to address the above-mentioned challenges, we have fabricated single-crystalline GeSn flexible PDs. Then, uniaxial tensile and compressive stresses were applied on the Ge/GeSn NMs by bending fixtures and its strain values were measured by Raman spectroscopy. We have investigated the effect of the uniaxial stress on photocurrent of the PDs and absorption coefficient of Ge/GeSn NMs experimentally and theoretically. Furthermore, change in responsivity by external strain was observed, proving the concept of light absorption modulation in flexible GeSn PDs.

Experiment

Figure 1. (a) The schematic fabrication process of a GeSn MSM PD: (i) GeSnOI substrate formed by direct wafer bonding and Si backside etching. (ii) Patterning NMs with array of etching holes followed by HF undercut process. (iii-iv) Pick up NMs by a PDMS stamp and then transfer-print it on PET substrate coated with SU8-2002. (v) Formation of rectangular GeSn mesas and interdigitated metal electrodes by dry etching and e-beam evaporation, respectively. (vi-vii) Bending GeSn MSM PD to apply uniaxial tensile and compressive strains. (b) Corresponding cross-sectional structure at each fabrication step. (c) An optical image of finished GeSn MSM PD and its zoomed-in microscopic image.

GeSn on insulator (GeSnOI) substrates were fabricated by a combination of direct wafer bonding and backside etching of Si.¹⁸ In short, a 90 nm unintentionally doped GeSn with a Sn composition of 8% was pseudomorphically grown on a Si substrate with a 1-μm-thick fully strain-relaxed unintentionally doped Ge buffer layer by CVD. A 500 nm sacrificial buried oxide layer (BOX) of SiO₂ was deposited on the GeSn surface by plasma enhanced chemical vapor deposition (PECVD), followed by chemical mechanical polishing (CMP). After direct bonding, the backside bulk Si was removed by tetramethylammonium hydroxide (TMAH) etching. The schematic of cross-sectional structure and its corresponding thicknesses of the GeSnOI were shown in Figure 1(a-i) and Figure 1(b-i), respectively. GeSnOI had a multiple layer of top 110 nm Ge, middle 90 nm GeSn, and bottom 300 nm SiO₂ formed on top of Si substrate as shown in Figure 2(a).

Figure 1(a) shows the fabrication process of a GeSn MSM PD. First, GeSnOI and polyethylene terephthalate (PET) substrates were thoroughly cleaned by acetone, IPA, and DI water. Standard

photolithography and dry etching were adopted to make NM patterns with etching holes (diameter: 3 μm and distance: 50 μm) on GeSnOI, followed by chlorine (Cl_2) gas-based reactive ion etching (RIE) to etch down the SiO_2 layer. Ge/GeSn NMs were then released by hydrofluoric acid (HF, 49%) undercut process (Fig. 1(a-ii)). Released Ge/GeSn NMs were transfer-printed on SU-8 2002 coated 175 μm PET substrates (Fig. 1(a-iii) and Fig. 1(a-iv)). Figure 1(b) shows its corresponding schematic cross-sectional structures at its respective steps. Rectangular-type Ge/GeSn NM mesas (size: $90 \times 100 \mu\text{m}^2$) were formed by RIE. Then, interdigitated metal pads (width and length of 4 and 100 μm , respectively) of Ti/Au (10/80 nm) were deposited on the mesas to finalize the device fabrication (Fig. 1(a-v)). Devices were conformally placed on bending fixtures along $\langle 100 \rangle$ direction with radii of 20 and 35 mm for bend-up and bend-down conditions, respectively (Fig. 1(a-vi, a-vii)).

Cross-sectional layer structure of the GeSnOI was inspected by field emission scanning electron microscopy (Apreo-S FESEM). Strain values applied in Ge/GeSn NMs were investigated by Raman spectroscopy (UHT S300 & WITEC) with a 532 nm laser focusing on a 1 μm diameter spot by a $100\times$ objective lens. Dark and photocurrent-voltage characteristics of the strained devices were characterized by semiconductor parameter analyzer (Keithley 3920) and a 2 μm wavelength laser (Thorlabs-FPL2000) with a beam size of 20 μm . Modulated bandgap and absorption coefficient of the strained GeSn were estimated by a theoretical model.

Results and Discussion

Figure 2. (a) Cross-sectional SEM image of GeSnOI. (b) Raman spectra of bulk Ge, GeSnOI, flexible GeSn NM under flat, bend-down, and bend-up conditions. The solid lines represent Lorentzian fits of the experimental data for determination of peak values.

Figure 2(b) shows the Raman spectra of bulk Ge, GeSnOI, Ge/GeSn NM on PET under flat, bend-down, and bend-up conditions, respectively. Raman peak values by the Ge-Ge vibration modes were obtained by Lorentzian functions and summarized in Table 1. It should be noted that Raman signal came from top Ge layer because penetration depth in Ge is around 20 nm when using a 532 nm laser. However, it is reasonable to claim that the collected Raman signal can represent the strain condition in GeSn because Ge/GeSn is pseudomorphically grown and the degree of bending is small. The peak of GeSnOI was measured at 299.73 cm^{-1} , which was 0.83 cm^{-1} red-shifted compared to the reference bulk Ge at 300.56

cm^{-1} . The peak of flat Ge/GeSn NM on PET was measured at 297.83 cm^{-1} , which was 2.73 cm^{-1} red-shifted compared to the reference bulk Ge at 300.56 cm^{-1} . From the Raman shifts, biaxial compressive strain in the GeSn layers was evaluated at around 1.12 and 0.83% for GeSnOI and Ge/GeSn NM, respectively.²⁴ Observed partial strain relaxation should be due to the undercut process. For the Ge/GeSn NM under bending-down condition, the peak was further red-shifted to 297.22 cm^{-1} , proving evidences of additional uniaxial tensile stress. Conversely, Raman peak was blue-shifted to 298.62 cm^{-1} in the bend-up condition, indicating that uniaxial compressive strain was applied by mechanical bending.

The mechanically applied uniaxial strain under bending conditions can be extracted by equation (1),²⁵

$$\varepsilon_{\text{mech}} (\%) = a \times \Delta\omega + b \times \Delta\omega^2 \quad (1)$$

where $\varepsilon_{\text{mech}}$ is the strain induced by mechanical bending along the $\langle 100 \rangle$ direction and $\Delta\omega$ is the Raman peak shift compared with that measured under flat condition. The coefficients a of 0.68 cm and b of 0.019 cm^2 are taken from those of Ge, which is a good approximation because the Sn content is not high. Strains applied by bending-down and bending-up were calculated to be 0.42% and -0.53%, respectively.

Table. 1. Raman peak values and extracted mechanical strains

Sample	Peak (cm^{-1})	$\Delta\text{Peak} (\text{cm}^{-1})$	$\varepsilon_{\text{mech}}$
Bulk Ge	300.56 ± 0.02	0	0
GeSnOI	299.73 ± 0.02	-0.83	0
GeSn/PET	297.83 ± 0.02	-2.73	0
Bend down	297.22 ± 0.02	-3.34	0.42%
Bend up	298.62 ± 0.02	-1.94	-0.53%

Figure 3. Band diagram of GeSn MSM PD

Figure 3 shows a band diagram of the GeSn MSM PD. Although both Ge and GeSn are unintentionally doped, they indicate a p-type background doping due to the supersaturated vacancy concentration originated from low growth temperature.²⁶ Typical p-type background doping

concentrations of Ge and GeSn are around 1×10^{16} and $1 \times 10^{17} / \text{cm}^3$, respectively.^{26,27} Depletion width in Ge/GeSn NM contacted with Ti/Au contact can be calculated by the following equation:

$$W = \left[\frac{2\varepsilon V_0}{q} \left(\frac{1}{N_a} \right) \right]^{1/2}$$

where ε is a permittivity of Ge/GeSn, V_0 is an energy barrier, q is an electron charge, and N_a is a doping concentration of Ge/GeSn.

For the doping concentrations of 1×10^{16} and $1 \times 10^{17} \text{ cm}^{-3}$, depletion widths are calculated to be 560 and 180 nm, respectively. Therefore, the depletion width entirely covers both Ge and GeSn, where the incident light is absorbed in GeSn. Incident $2 \mu\text{m}$ wavelength photons are not absorbed by top Ge, but are absorbed mostly by bottom GeSn layer. Absorption coefficient (α) of Ge maintains its high values in ultraviolet (UV), visible (VIS), and short wave NIR region. However, it sharply drops near $1.55 \mu\text{m}$ due to the direct band gap energy of 0.8 eV. Therefore, both quantum efficiency and responsivity of typical Ge PDs sharply drop when the wavelength of incident light becomes larger than $1.55 \mu\text{m}$. α of Ge at $2 \mu\text{m}$ is as close as 1 cm^{-1} ,²⁸ indicating the light absorption by Ge is negligible at this wavelength. In addition, a depletion region is formed in both Ge and GeSn, leading to an efficient generation of photo-carriers in GeSn. Therefore, the absorption solely occurs in GeSn.

Figure 4. (a) Dark (black line) and photocurrent (red line) versus applied voltage bias under flat (solid), bend-up (short dotted line), and bend-down (long dotted line) conditions. (b) Calculated absorption spectra of GeSn. (c) Photocurrent with different optical powers at 2 V. (d) Responsivity at 2000 nm with different mechanical strains.

Current-voltage characteristics of the flexible GeSn MSM PDs were measured under dark and illuminated conditions. Figure 4(a) shows dark and photocurrent of the device at a bias range from 0 to 2 V under flat, bend-down, and bend-up conditions. Dark current was increased from 8.1 to 10.3 μA in the bend-down (i.e., uniaxial tensile stress) condition while it was decreased to 7.2 μA in the bend-up (uniaxial compressive stress) condition at 2 V. Compared with other reported dark current densities of GeSn PDs (i.e., from around 0.1 to 10 A/cm^2),^{9,29} dark current density of our flat PD was measured to be as low as 0.09 A/cm^2 at 2 V, indicating the excellent quality of the transfer-printed Ge/GeSn NMs. Increased dark current under the tensile strain is due to increased electric field by the shrinkage of direct bandgap of the strained GeSn.³⁰ Higher degree of increased dark current was observed under tensile strain compared to that of reduced dark current under compressive strain. The major component of the dark current is the carrier injection over Schottky barrier height (SBH). Though SBH is reduced in tensile strain by shifting up the valence band with light hole on top, the reduction in SBH represents only 1% of SBH between metal electrodes and Ge.³¹ Therefore, the change in dark current of the strained device should be attributed to strain-dependent modulation of bandgap of GeSn. Detailed analysis of bandgap modulation will be discussed in a later part of this paper.

Photocurrent of the flat and strained devices was measured using a 2 μm laser with an incident optical power of 23.21 mW. Photocurrent at 2V increased from 17.4 to 43.6 μA while it decreased to 11.3 μA under bend-down and bend-up conditions, respectively. More precisely, 0.42% tensile strain increased the photocurrent by 151%, while 0.53% compressive strain decreased the photocurrent by 35%. This trend is in accordance with the strain-dependent modulation of bandgap and its resultant absorption coefficient. The strained bandgap energies and absorption coefficient were calculated by using the deduced strain values and the theoretical models.³² Figure 4(b) shows the calculated absorption spectra for the flat and strained Ge/GeSn NMs as a function of wavelength. The photodetection cutoff wavelength of bulk Ge PDs is around 1.55 μm due to the direct bandgap of 0.8 eV. 8% Sn alloying increases the cutoff wavelength to 2077 nm (597 meV). Mechanical strain further modifies the band structure of the strained Ge/GeSn NMs. Our calculations indicate that the direct bandgap was shrunked to 583 meV and the absorption coefficient at 2000 nm was increased from 2211 to 2655 cm^{-1} by 0.42% tensile strain. In contrast, the bandgap was increased to 607 meV and the absorption coefficient was reduced to 1546 cm^{-1} at 2000 nm by 0.53% compressive strain. These calculations support that the modulation of photocurrent is attributed to the strain-induced changes in the absorption coefficient. It also indicates that bandgap changes more under tensile strain (e.g. 14 meV V.S. 10 meV), which agreed well with the other result.³³

Figure 4(c) shows the measured photocurrent at 2000 nm with incident optical powers of 1.19, 5.21, 15.46, and 23.21 mW for the GeSn PDs under flat and strained conditions. The photocurrent increased linearly with increasing optical power, indicating that photo-generated carriers are the major component of the photocurrent. To further investigate the effect of mechanical bending on the performance of PDs, responsivity was extracted as shown in Figure 4(d). The responsivity significantly

increased from 1.03 to 3.68 mA/W with 0.42% tensile strain, while it decreased to 0.45 mA/W with 0.53% compressive strain.

Conclusion

In summary, we demonstrated flexible GeSn MSM PDs and investigated the strain effect on the device performance via theoretical and experimental demonstration. The devices were fabricated from the transfer-printed Ge/GeSn NMs on PET substrates. Raman spectroscopy was used to estimate induced uniaxial tensile strain of 0.42% and compressive strain of 0.53% under bend-down and bend-up conditions, respectively. Mechanical strain modulated bandgap energy and absorption coefficient of GeSn, leading to changes in dark current, photocurrent, and responsivity of strained GeSn PDs. Dark current was increased from 8.1 to 10.3 μ A at 2 V by tensile strain while it was decreased to 7.2 μ A by compressive strain. Photocurrent was increased from 17.4 to 43.6 μ A while it was decreased to 11.3 μ A by tensile and compressive strain, respectively. Relative changes in the responsivity were also observed from strained GeSn PDs, which indicated the changes in the absorption coefficient during bending. In addition, larger changes in current and responsivity were observed under smaller tensile strain. The different amount of changes should be ascribed to asymmetrical changes of bandgap under tensile and compressive strain. The study indicates that GeSn NMs can be used to realize flexible NIR PDs with an absorption wavelength at 2 μ m.

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