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Transfer-printing-enabled GeSn flexible resonant-cavity-enhanced photodetectors with strain-amplified mid-infrared optical responses†

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Mid-infrared (MIR) flexible photodetectors (FPDs) constitute an essential element for wearable applications, including health-care monitoring and biomedical detection. Compared with organic materials, inorganic semiconductors are promising candidates for FPDs owing to their superior performance as well as optoelectronic properties. Herein, for the first time, we present the use of transfer-printing techniques to enable a cost-effective, nontoxic GeSn MIR resonant-cavity-enhanced FPDs (RCE-FPDs) with strain-amplified optical responses. A narrow bandgap nontoxic GeSn nanomembrane was employed as the active layer, which was grown on a silicon-on-insulator substrate and then transfer-printed onto a polyethylene terephthalate (PET) substrate, eliminating the unwanted defects and residual compressive strain, to yield the MIR RCE-FPDs. In addition, a vertical cavity was created for the GeSn active layer to enhance the optical responsivity. Under bending conditions, significant tensile strain up to 0.274% was introduced into the GeSn active layer to effectively modulate the band structure, extend the photodetection in the MIR region, and substantially enhance the optical responsivity to 0.292 A W^{-1} at $\lambda = 1770 \text{ nm}$, corresponding to an enhancement of 323% compared with the device under flat conditions. Moreover, theoretical simulations were performed to confirm the strain effect on the device performance. The results demonstrated high-performance, nontoxic MIR RCE-FPDs for applications in flexible photodetection.

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Introduction

Flexible optoelectronic devices operating in the mid-infrared (MIR) region—generally defined in the spectral range of $1.8\text{--}5 \mu\text{m}^1$ —have received considerable attention for use in a wide range of emerging applications such as gas sensing, strain sensing, optical links, environment monitoring, health diagnosis, and wearable photonic textiles.^{2–4} Among the various components required for flexible optoelectronic systems, photodetectors (PDs) are crucial for optical – electric conversion. Although flexible PDs (FPDs) composed of organic or 2D materials operating in the MIR region have been extensively explored,^{5–8} they typically suffer from low quantum efficiency

resulting from weak absorption and poor reliability. Meanwhile, semiconductor-based FPDs are being extensively used owing to their unique advantages of superior quantum efficiency.^{9–12} To achieve efficient MIR photodetection, narrow-bandgap semiconductors are necessary. Currently, compound semiconductors based on III–V materials such as InAs and InSb that are dominating the market satisfy this requirement, and FPDs based on III–V materials have been demonstrated as well.¹² However, the toxicity and wafer cost hinder the practical applications.

Moreover, FPDs based on group-IV semiconductors are being explored for MIR photodetection, because they are nontoxic, environment-friendly, and affordable, thereby rendering them more suitable for wearable applications. Although the applicability of Si^{13,14} and Ge^{15–17} PDs has been demonstrated, their application to MIR photodetection is difficult owing to the cut-off wavelength of 1.1 and 1.5 μm , respectively. To extend the photodetection range to the MIR region, tensile strain can be employed using various approaches such as thermal expansion gradient between Si and Ge^{18,19} and stressors;²⁰ however, a limited amount of tensile strain can be incorporated which creates challenges. Another promising approach is alloying Ge with Sn, which is another group-IV element.^{21,22} With the addition of Sn into Ge, the direct

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bandgap can be effectively reduced, thereby enabling efficient absorption in the MIR region.^{23–25} Moreover, the presence of L-conduction band in GeSn alloys generates a unique momentum – space carrier separation scheme,^{25,26} which facilitates high-performance MIR photodetection. Recent theoretical studies have reported that the performance of GeSn PDs is comparable, and can be even superior to that of the current commercially available semiconductor MIR PDs,^{24,25} which make it a promising candidate for low-cost practical applications. Despite the limited solubility of Sn in Ge of ~1%,^{27,28} the recent advancements in low-temperature growth techniques using molecular beam epitaxy (MBE),^{27,28} chemical vapor deposition (CVD),^{29–31} and sputter³² have enabled the growth of high-quality GeSn layers on Si or silicon-on-insulator (SOI) substrates, which can yield Sn contents up to 36%.³⁰ Various GeSn-based PDs have been demonstrated with an extended photodetection range,^{33–48} attaining up to 3.7 μm ⁴⁷ and enhanced quantum efficiency, which highlights their potential for efficient MIR photodetection. However, the epitaxy of GeSn layers on Si or SOI substrates typically requires an appropriate buffer (usually Ge) to improve the material quality and induces an indispensable biaxial compressive strain.³⁵ This unfavorable biaxial compressive strain significantly increases the bandgap energies and weakens the effectiveness of Sn-alloying, thereby hindering the performance of GeSn-based PDs.^{23,48} A few approaches have been proposed to eliminate biaxial compressive strain, such as suspended structures,⁴⁹ graded GeSn buffer technologies,⁴⁰ and wafer-binding technologies,⁵⁰ and an extended photodetection range up to 4.6 μm has been experimentally achieved.⁴⁹ However, as these GeSn PDs are fabricated on rigid substrates, they lack the flexibility required for wearable applications.

To the best of our knowledge, this study demonstrates the first strain-free GeSn MIR resonant-cavity-enhanced FPDs (RCE-FPDs), wherein the performance is further enhanced under bending conditions. In particular, single-crystal GeSn nanomembranes (NMs) were grown on an SOI substrate using low-temperature MBE and subsequently transferred on to flexible polyethylene terephthalate (PET) substrates. The transfer was performed using a transfer-printing technique with a mechanically strong SU8 adhesion layer to eliminate unwanted defective region and residual compressive strain. Thus, a device with improved performance was realized. After the transfer, GeSn active layer sandwiched between a low-refractive-index (RI) SU8 layer and air creates a vertical cavity enabling a multiple-pass reflection scheme, which enhances the light-matter interaction under resonance conditions and significantly augments the optical responsivity. Notably, if the GeSn RCE-FPDs are subjected to bending conditions, a tensile strain up to 0.274% can be introduced into the GeSn active layer. This tensile strain can effectively modulate the band structure, extend the photodetection range, and significantly enhance the absorption in the MIR region. Furthermore, the strain effect on the device performance was theoretically analyzed to support the experimental findings. The results demonstrate the feasibility of GeSn RCE-FPDs with enhanced optical

responses, establishing a new avenue for high-performance, nontoxic, sensitive, reliable, and flexible MIR photodetectors for versatile wearable applications.

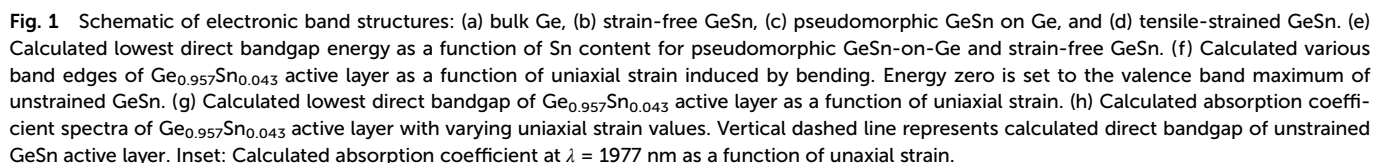
Analysis of impact of strain on band structure of GeSn alloys

For FPDs, a strain can be induced under the bending condition, which will significantly impact the device performance. We first systematically investigated the impact of strain on the electronic band structure, optical absorption, and device performance of GeSn PDs. The direct bandgap of bulk Ge is 0.7985 eV at $T = 300$ K (Fig. 1(a)). By adding Sn into Ge, the electronic band structure can be effectively modified (Fig. 1(b)). As a result, the direct-gap absorption edge can be red-shifted to extend the photodetection range and enhance the quantum efficiency of GeSn PDs. Currently, a suitable buffer (typically Ge) is used to facilitate the growth of GeSn on Si substrates and improve the material quality. However, as shown in Fig. 1(c), the growth of $\text{Ge}_{1-x}\text{Sn}_x$ layer on Ge buffer within a critical thickness is pseudomorphic,³⁵ leading to a significant in-plane biaxial compressive strain of 0.143x owing to the larger lattice constant of GeSn than Ge. This compressive strain lifts the direct conduction band and shifts the heavy-hole (HH) band above the light-hole (LH) band. As a result, the lowest direct-gap transition takes place between the HH band and Γ -valley conduction band, which increases with increasing compressive strain. Fig. 1(e) shows the calculated lowest direct-gap transition energy for bulk GeSn and pseudomorphic GeSn-on-Ge as a function of Sn content. Evidently, the compressive strain can significantly increase the lowest direct-gap transition energy, equivalently reducing the “effective Sn content” in the GeSn active layer. Thus, it is essential to eliminate the unwanted compressive strain to achieve strain-free GeSn alloys and “recover” the performance of GeSn PDs. Furthermore, the performance of GeSn PDs can be further enhanced by introducing tensile strain/stress to modify the energy band (Fig. 1(d)). Here we investigate the band structure of GeSn subjected to a uniaxial stress (σ) along $\langle 100 \rangle$ direction induced by bending, as shown in the inset in Fig. 1(f). The resulting strain fields can be expressed as

$$\begin{aligned}\epsilon_{xx} &= \epsilon_a \\ \epsilon_{yy} &= \epsilon_{zz} = -(C_{12}/C_{11})\epsilon_a \\ \epsilon_{xy} &= \epsilon_{yz} = \epsilon_{zx} = 0\end{aligned}\quad (1)$$

where C_{11} and C_{12} denote the elements of stiffness matrix of GeSn, which were obtained through the linear interpolation between those of Ge and Sn.²² ϵ_a represents the normal strain along $\langle 100 \rangle$ direction. The band structure was evaluated using a multiple-band k - p method, considering the band nonparabolicity effect^{23,51} and the strain-induced energy shifts were modeled using the deformation potential theory.⁵¹ Thereafter, the absorption coefficient was calculated using Fermi's golden rule.⁵¹ The parameters used in the simulations were obtained





2.89 eV denotes the bowing parameter for GeSn alloys at $T = 300$ K.^{23,35} The calculated band edges and the lowest direct bandgap of the GeSn layer formulated as a function of uniaxial strain are presented in Fig. 1(f) and (g), respectively. With the application of the compressive strain (*i.e.*, under bend-up conditions), the Γ -conduction band shifted to a higher energy. Moreover, for the valence band, the HH shifted above the LH

band (Fig. 1(f)). Consequently, the lowest direct bandgap was defined as the transition energy from the HH band and the Γ -conduction band ($\text{HH} \rightarrow \text{c}\Gamma$), which increased with the increasing compressive strain, as shown in Fig. 1(g). By contrast, with the application of the tensile strain (*i.e.*, under bend-down conditions), the Γ -conduction band shifted to a lower energy, whereas the LH band shifted above the HH band (Fig. 1(f)). Thus, the lowest direct bandgap can be defined as the optical transition from the LH band to the Γ -conduction band ($\text{LH} \rightarrow \text{c}\Gamma$), and it decreased with the increasing tensile strain (Fig. 1(g)). The calculated direct-gap absorption coefficient spectra of the GeSn active layer with various uniaxial strains are shown in Fig. 1(h). Without strain, the absorption coefficient decreased with the increasing wavelength and becomes negligible at the direct bandgap. With the application of the compressive strain, the lowest bandgap significantly increased, thereby blue-shifting the direct-gap absorption edge. Thus, the absorption coefficient reduced as it is proportional to the joint density-of-state (JDOS; for bulk materials, the JDOS is proportional to $\sqrt{\hbar\omega - E_g}$ ⁵¹). By contrast, the application of the tensile strain reduces the direct bandgap, thereby red-shifting the absorption spectrum, increasing the JDOS, and significantly enhancing the absorption coefficient. The calculated absorption coefficient at $\lambda = 1977$ nm is depicted as a function of uniaxial strain in the inset of Fig. 1(h). The absorption coefficient is enhanced by a factor of 301% for the increase in strain from 0% to 0.274%, thereby enhancing the quantum efficiency of the PDs.

Material growth and device fabrication

The fabrication of the GeSn RCE-FPD involves the growth and fabrication of the material. In this study, the sample was prepared using low-temperature MBE on an SOI substrate with a 2.5 μm -thick top Si layer and a 1 μm -thick buried oxide (BOX) layer under a base pressure less than 5×10^{-11} torr. The epitaxy started with the growth of a strain-relaxed Ge virtual substrate (VS) using a two-step growth technique: (a) a 100 nm-thick Si buffer layer grown at 600 $^{\circ}\text{C}$, (b) a 100 nm-thick Si buffer layer grown at 350 $^{\circ}\text{C}$, (c) a 60 nm-thick Ge seed layer grown at 350 $^{\circ}\text{C}$ followed by *in situ* thermal annealing at 800 $^{\circ}\text{C}$ for 5 min, and (d) a 60 nm-thick Ge layer grown at 550 $^{\circ}\text{C}$. To suppress Sn segregation, the growth temperature was reduced to 150 $^{\circ}\text{C}$ to grow a 370 nm-thick intrinsic GeSn layer. The structure was capped with a 3 nm-thick Si layer. Note that although the intrinsic GeSn layer was undoped, a background *p*-type doping concentration of $6.5 \times 10^{16} \text{ cm}^{-3}$ was detected by Hall measurements. The cross-sectional transmission electron microscopy (XTEM) image of the grown sample is illustrated in Fig. 2(a), wherein significant misfit dislocations that distinguish the interface between the Ge and top Si layer were observed. However, they were absent in the GeSn active layer. By contrast, a sharp and flat interface was observed between the Ge VS and GeSn active layer, as shown in the lower inset of Fig. 2(a). A selected electron diffraction of

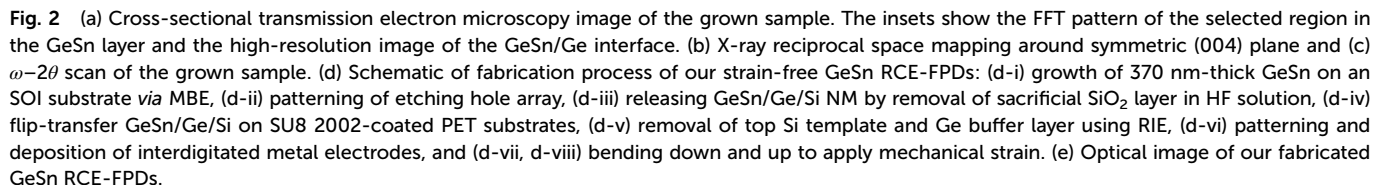
the GeSn active layer, as shown in the higher inset of Fig. 2(a), confirmed the single-crystalline nature and desired quality of the grown sample material. Additionally, X-ray diffraction (XRD) techniques were employed to probe the structural properties of the grown sample. The X-ray reciprocal space mapping (XRDSM) of symmetrical (004) plane is presented in Fig. 2(b), which portrays two distinguished peaks that can be ascribed to the Ge and GeSn layers, respectively. Note that the two peaks were vertically aligned with the same Q_x value, implying that the GeSn layer was pseudomorphic to the underlying Ge VS. The high-resolution ω - 2θ scan of the grown sample is presented in Fig. 2(c), from which the Bragg's angles of the Ge and GeSn layers were extracted to determine their lattice constants along the growth direction. According to the analysis procedure described in ref. 41, the GeSn layer had Sn content of 4.3% and an in-plane biaxial compressive strain of 0.52%.

The procedure for fabricating the GeSn RCE-FPDs is illustrated in Fig. 2(d). The process initiated with an epitaxial growth of 370 nm-thick GeSn on a SOI with a Ge VS using MBE (Fig. 2(d-i)). Subsequently, the photoresist was first spin coated to protect the GeSn layer and for lithography. An array of holes with a diameter of 3 μm and a spacing of 50 μm were patterned with standard optical lithography and etched down to expose the sacrificial layer (BOX layer) by reactive ion etching (RIE) (Fig. 2(d-ii)). Thereafter, the samples were placed in 49% hydrofluoric acid (HF) solution for 30 min to release the GeSn/Ge/Si NMs by removing the sacrificial layer (Fig. 2(d-iii)). The released GeSn/Ge/Si NMs were flip transferred on SU8 2002-coated PET substrate with a thickness of 175 μm (Fig. 2(d-iv)). Subsequently, the top Si and Ge layers on the GeSn were etched away using RIE to alleviate the unwanted residual compressive strain caused by material growth and achieve strain-free GeSn NMs (Fig. 2(d-v)). Further, to remove the defective region in the GeSn layer and improve the device performance, the GeSn layer was over-etched, and the final thickness of GeSn NMs was estimated as 240 nm. Eventually, Ti/Au (10/80 nm) interdigitated pads (length: 100 μm ; width and pitch: 4 μm) were deposited using an e-beam evaporator to complete the fabrication of the devices (Fig. 2(d-vi)). An optical image of the fabricated GeSn RCE-FPD is displayed in Fig. 2(e), which evidently demonstrates the flexibility of the device. Thereafter, the devices were conformally attached along $\langle 100 \rangle$ direction with extreme precautions on the convex (Fig. 2(d-vii)) and concave (Fig. 2(d-viii)) bending fixtures with a radius of R to introduce uniaxial stress under bend-up and bend-down conditions, respectively. For comparison, a GeSn reference PD was fabricated using the as-grown GeSn on the SOI substrate with the same device structure (labelled as GeSn-on-SOI device).

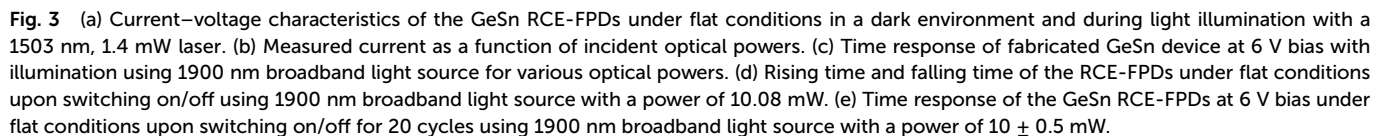
Electrical characterization

The current-voltage characteristics of the fabricated GeSn RCE-FPDs under flat conditions in a dark environment and





illumination, the photoresponses were recorded with clear on/off switching behavior with a good on/off ratio of ~ 40 . It is noted that the obtained on/off ratio is lower than the typical values of 10^2 – 10^5 for 2D-material-based MIR PDs^{52,53} and $\sim 10^8$ for InGaAs/InP *p-i-n* FPD,¹² mainly attributed to the relatively high dark current of the metal–semiconductor–metal PD structure.⁵⁴ By suppressing the dark current, it is possible to further enhance the on/off ratio to improve the device performance. With increased optical power, the photocurrent increased accordingly. Fig. 3(d) shows the time response of the GeSn RCE-FPD upon switching on/off with the rising time (t_r) and falling time (t_f), which denote the time required for the photocurrent to increase from 10% to 90% of the maximum and decreases from 90% to 10% of the maximum photocurrent, respectively. Our GeSn RCE-FPDs exhibited short rising time and falling times of $t_r = 1.27$ ms and $t_f = 1.14$ ms, respectively. The fast response time is desirable for flexible and wearable devices. The response time of metal–semiconductor–metal PDs is mainly limited by the transit time of photogenerated carriers which related to the interelectrode spacing.⁵⁵ Note that high-speed GeSn PDs with a bandwidth of 30 GHz has been experimentally demonstrated.⁵⁶ Thus, it is anticipated that further optimization of our GeSn RCE-FPDs could significantly improve the response time to achieve high-speed operation.^{57,58} Fig. 3(e) shows the

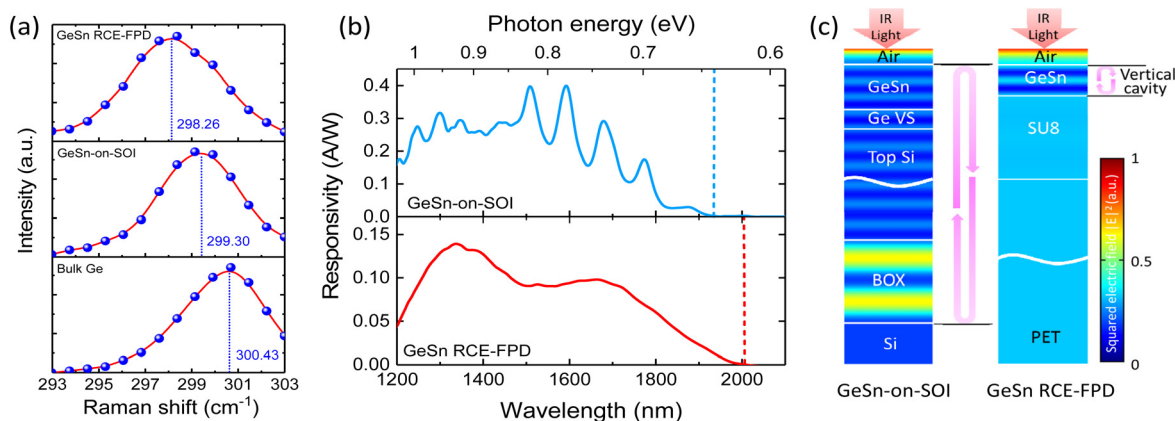


ditions, the peak further shifted to 298.26 cm^{-1} . The Raman shift is related to strain *via* the expression $\Delta\omega = k \times \varepsilon$, where the strain-shift coefficient for GeSn is $k = -181\text{ cm}^{-1}$ from ref. 59. Thus, the shift of Raman peak to a lower wavenumber is an indication of relaxation of the compressive strain in the GeSn layer.

The optical responsivity of the devices was measured using a tunable light source comprising a 250 W quartz tungsten halogen (QTH) lamp and a 25 cm monochromator. After the emitted light was optically chopped at 200 Hz, it was incident normally on the device using an objective of 50 $\times$. A source-meter (Keithley 2400) was used to provide voltage to the device in series with a load resistance of 50 Ω . The voltage drop across the load resistance was accurately recorded using a lock-in amplifier (SRS, SR830) to determine the photocurrent. A lock-in technique was adopted to enhance the signal-to-noise ratio. The incident optical power on the device was measured using an optical powermeter with an E-InGaAs photodetector (Thorlabs, PM 400 & S148C) to determine the optical responsivity. The room-temperature optical responsivity spectra of the GeSn-on-SOI PD and GeSn FPD under flat conditions at 6 V bias voltage are presented in Fig. 4(b). Unlike typical smooth responsivity curves in GeSn PDs,³⁵ the responsivity spectrum of the GeSn-on-SOI PD exhibited several peaks with a free-spectrum-range (FSR) of ~ 90 nm, which can be primarily attributed to the vertical cavity effect. The responsivity reduced with an

Optical characterization

First, we compared the optical properties of the GeSn RCE-FPD and GeSn-on-SOI PD. Raman analysis was performed to characterize the strain status of the samples using a Raman microscopy (UHT S300 & WITEC) with a 532 nm excitation laser under backscattering scheme. The laser light was focused on the sample surface using an objective of 100 $\times$ and a spot size of ~ 1 μm . The measured Raman spectra of the bulk Ge, as-grown GeSn-on-SOI, and flexible GeSn RCE-FPD under flat conditions are presented in Fig. 4(a). After fitting the experimental data using Gaussian functions, the peak positions were determined. For bulk Ge, the Raman peak corresponding to the longitudinal phonon-phonon mode appeared at 300.43 cm^{-1} . For the GeSn-on-SOI, the Raman peak shifted to 299.30 cm^{-1} . This shift was attributed to the introduction of Sn into the material. For the GeSn RCE-FPD under flat con-



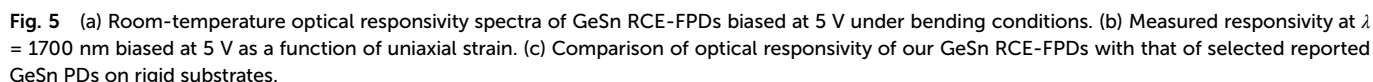
increase in the wavelength and became negligibly small at ~ 1930 nm, corresponding to the direct bandgap of the material (0.642 eV). By contrast, for the GeSn RCE-FPD, the responsivity spectrum exhibited two peaks at 1339 and 1666 nm, corresponding to an FSR of 327 nm, suggesting the existence of resonant cavity and the effect of the substrate (ESI[†]). In addition, the responsivity decreased with increasing wavelength until the cutoff at ~ 2030 nm, corresponding to the direct bandgap of the material (0.610 eV). The extension of the cutoff wavelength could be attributed to the relaxation of 0.52% compressive strain initially stored in the GeSn active layer that significantly reduced the bandgap energy. In addition, we noted that the small responsivity of GeSn RCE-FPD compared to that of the reference GeSn-on-SOI PD could be primarily because of the thinner active layer (240 nm for the GeSn RCE-FPD and 360 nm for the reference GeSn-on-SOI PD). Further, to confirm the vertical cavity effect, simulations based on finite element method (FEM) were performed to derive the optical field distributions for the GeSn-on-SOI PDs and GeSn RCE-FPDs. For the FEM simulations, the wavelength-dependent RIs of the materials were obtained from ref. 60 and 61. A plane wave was employed as the light source with a normal incidence onto the devices, and subsequently, the field distributions were obtained. The optical field simulated for the devices under the resonant conditions is depicted in Fig. 4(c). For the GeSn-on-SOI PDs, the incident light entering the device experiences reflections at the air/GeSn and BOX/Si interfaces owing to the substantial difference between the RIs ($n(\text{GeSn}) = 4.23$, $n(\text{SiO}_2) = 1.45$). Consequently, a Fabry–Perot cavity was created, as evident from the standing pattern. By contrast, for the GeSn RCE-FPD, adequate reflections were observed at the GeSn/air and GeSn/SU8 interfaces because of the large contrast in the RI ($n(\text{GeSn}) = 4.23$, $n(\text{SU8}) = 1.562$). Thus, a vertical cavity was created to enhance the light–matter interaction and responsivity. In addition, the cavity length of the GeSn RCE-FPD was smaller than that of GeSn-on-SOI PD, resulting in a larger FSR that is consistent with the experimental observation.

Thereafter, we investigated the optical responses of the GeSn RCE-FPDs under flat, bend-down, and bend-up conditions along $\langle 100 \rangle$ direction. The uniaxial strain induced by bending can be evaluated as⁶²

$$\epsilon_a = -\frac{L}{R}, \quad (3)$$

$$\varepsilon_a = -\frac{L}{R}, \quad (3)$$

where $L \sim 83 \text{ }\mu\text{m}$ denotes the distance from the neutral plane of the device to the GeSn film of the GeSn RCE-FPD. Based on the analysis, we can determine the strain induced by mechanical bending, *e.g.*, the tensile (compressive) strain was induced under bend-down (bend-up) conditions. The measured responsivity (R_λ) spectra of the GeSn RCE-FPD under flat and bending conditions are plotted in Fig. 5(a). Compared with the case of the flat condition, the optical responsivity significantly increased under the bend-down condition owing to the increasing tensile strain. In addition, the cutoff wavelength was slightly extended to a longer wavelength with increasing tensile strain. On the contrary, the optical responsivity significantly decreased under bend-up conditions, and the cutoff wavelength blue-shifted with the increasing compressive strain. In Fig. 5(b), the optical responsivity at $\lambda = 1770 \text{ nm}$ is portrayed as a function of the uniaxial strain, and the corresponding enhancement factor is defined as $E_F = R_\lambda(\varepsilon_a)/R_\lambda(\varepsilon_a = 0)$. With the application of the tensile strain, the responsivity increased significantly, and a notable enhancement factor up to 323% for $\varepsilon_a = 0.274\%$ was achieved, thereby validating sensitive photodetection. To demonstrate the competitiveness of our GeSn RCE-FPDs, Fig. 5(c) shows the comparison of the optical responsivity at room temperature for our GeSn RCE-FPD with selected reported GeSn PDs on rigid substrates^{35,38,39,63–66}. It can be seen that, under flat condition ($\varepsilon_a = 0$), the optical responsivity of our GeSn RCE-FPDs is comparable with these GeSn PDs with Sn content of $<5\%$ and similar active layer thicknesses. With the application of $\varepsilon_a =$



review & editing. M. K. – Conceptualization, project administration, formal Analysis, writing – review & editing. G. E. C. – Conceptualization, supervision, project administration, formal analysis, writing – review & editing.

There are no conflicts to declare.

Conclusions

Acknowledgements

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Author contributions

Y. C. T. – Methodology, investigation, formal analysis, validation. A. S. – Methodology, investigation, formal analysis, validation. P. R. H. – Methodology, investigation. J. T. J. – Methodology, investigation. K. C. L. – Methodology, investigation. H. H. C. – Methodology, investigation, writing –

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