Large-area flexible Visible-Short wavelength Infrared Photodetection based on III-V compound semiconductor membrane photodetector with high performance

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Abstract

Mechanically flexible optoelectronic devices and systems can enable a much broader range of applications than what their rigid counterparts can do, especially for novel bio-integrated optoelectronic systems, flexible consumer electronics and wearable sensors. Inorganic semiconductor could be a good candidate for the flexible PD when it can keep its high performance under the bending condition. Here, we demonstrate a III-V material-based flexible photodetector operating wavelength from 640 nm to 1700 nm with the high detectivity of $5.18 \times 10^{11} \text{ cm}^2\text{Hz}^{1/2}/\text{W}$ @1550nm by using a simply top-to-down fabrication process. The optoelectrical performances are stable as the PDs are exposed to bending cycles with a radius of 15 mm up to 1000 times. Furthermore, the mechanical failure mode of the PD is also investigated, which suggests that the cracking and delamination failure mode are dominant in bending up and bending down direction, respectively. Such a flexible III-V material-based PD and design with stable and high performance could be a promising strategy for the application of the flexible broad spectrum detection.

Introduction

Emerging application of mechanically flexible optoelectronic devices have flourished in the fields of flexible imaging/displays¹⁻⁵, sensors⁶⁻⁸, short-reach data communications⁹⁻¹¹, solar cell¹²⁻¹⁴ and so on. As an indispensable component of flexible optoelectronic systems, flexible photodetectors (PDs) have been extensively studied and achieved breakthroughs in the kinds of response wavelength ranges based on several material platforms and one of the most important PDs is the visible-infrared broad PD. Organic PD is the one candidate for next-generation PDs due to their tunable light detection ranges, solution processability, low-cost, lightweight, mechanical flexibility, and seamless heterogeneous integration. A great progress has been achieved for visible or near IR organic PDs, but there are several key bottlenecks, such as large bandgap, weak absorption, and poor charge generation in the NIR region, resulting in poor performance¹⁵⁻¹⁷. Moreover, the inorganic semiconductor nanowires (NWs)¹⁸⁻²⁰, and 2D materials²¹ have also been applied in the flexible PDs and great progress have been achieved at the visible to short-wavelength IR range because of their unique electronic structure, easy fabrication, broad spectra absorption and flexibility. In the recent years, the devices based on graphene^{22, 23}, PS/graphene²⁴, and HgTe/Graphene²⁵ have been performed to fabricate the flexible broadband PDs. However, the commercial application is limited by their weak light absorption, large noise, high dark current, the stability and reproduction. As a result, those approaches are still some way from being a commercial solution.

Inorganic semiconductor membrane, as an ultrathin layer, is another strong candidate for the flexible PDs because of its excellent flexibility and bendability, while keeping their intrinsic crystal quality and optoelectrical properties. In the application of visible light to short wavelength infrared (SWIR) detection, flexible inorganic PDs have been implemented based on Si^{26, 27} and Ge^{28, 29}, but there are still some disadvantages for the broad-spectra high-performance PD. For example, Si membrane PDs are limited in visible wavelength-sensing range and low quantum efficiency due to the property of indirect band gap

closed to 1 eV, while Ge membrane flexible PD shows relatively high dark current attributed to recombination centers located in the damaged Ge. As we know, InGaAs/InP PD has been widely applied and commercialized in the night vision, inspection, agricultural sorting, and so on, due to its broad spectra and high-performance. Thus, in this work, combined with the mature material growth and device fabrication, we have demonstrated the flexible InGaAs membrane PD with the detectivity of $5.18 \times 10^{11} \text{ cm}^2 \text{ Hz}^{1/2} / \text{W}$ by lifting off epitaxial layers from their native substrates using a sacrificial layer and transferring them onto a flexible host carrier. A sidewall passivation is applied to simplify InGaAs PD fabrication process, and large-area flexible PDs with operating wavelength from 600 nm to 1700 nm are obtained. Furthermore, the optoelectrical performance of such flexible PDs keeps stable in bending state and the physical mechanism is also studied in details. As a III-V materials based flexible PD with the conventional high performance of the rigid PDs, one can envision its role for the operation from visible light to short wavelength IR in the novel bio-integrated optoelectronic systems, flexible consumer electronics and wearable sensors.

Methods

The flexible InGaAs PIN PD fabrication process is schematically illustrated in Fig. 1(a). The process combines constructing metal frame, etching out the release holes, fabricating the passivation layer, peeling off the membranes, depositing the bottom electrode contact and finally transferring the device layers to the flexible organic substrate. Details of the the flexible PD fabrication and characterization are as follows.

Material growth

Firstly, the III-V PIN PD layers were grown on the 2-inch InP substrate by metal organic chemical vapor deposition. The thickness for the p- and n-type InP layers were 600 nm and 500 nm, respectively. There were three lattice matched $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ layers in the structure, including the top 50-nm-thick p-InGaAs layer as the ohmic contact layer, the 2.2 μm -thick undoped InGaAs layer as the absorption layer, and the 500nm-thick n-InGaAs layer between n-InP contact layer and n-InP buffer layer is the sacrificial layer. It is noted that the p-type InGaAs layer and InP layer were achieved by single MOCVD diffusion of zinc after the epitaxy. Then, by using micro/nano fabrication technique, a metal frame layer (20 nm/200 nm Ti/Au) was deposited on the top of the device layers and thus two 150 μm -wide metal bars formed the metal frame layer. The metal frame layer can be used as the supporting frame to increase the mechanical strength of the to-be released InGaAs-based membrane and the top contact layer for the PD devices.

Device Fabrication

Arrays of $40 \times 40 \mu\text{m}^2$ square release holes inside were patterned on the device layers in $\sim 100^\circ$ and $\sim 10^\circ$ directions, by selectively wet etch down to the surface of the InGaAs sacrificial layer. The release holes were prepared to facilitate the transverse wet etching of InGaAs sacrificial layer beneath the device layer. In the process of selectively wet etching, a $\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2:\text{H}_2\text{O}$ (1:8:120) solution selectively corroded

InGaAs, but had no effect on InP. On the contrary, the mixed solution of $\text{HCl}:\text{H}_3\text{PO}_4$ (1:3) selectively etched InP but has no effect on InGaAs. Therefore, when InGaAs or InP were etched alternately with different etching solution, excessive etching could be properly performed to ensure sufficient corrosion. Based on the selective wet-etching process, the release holes were formed down to the surface of the InGaAs layer (sacrificial layer). The release holes in detail with scan electron microscopy (SEM) were shown in Fig. 2. As the etching rates of $\langle 100 \rangle$ and $\langle 010 \rangle$ direction were different³⁰, the periodic spacing between the release holes was designed to be 240 μm in $\langle 010 \rangle$ and 290 μm in $\langle 100 \rangle$, respectively, as shown in Fig. 2. Compared with the same spacing distances in each directions between the release holes^{31,32}, the different distances design ensured that the delamination could be completed simultaneously in both directions in the process of the horizontal wet etching separation. Furthermore, As can be seen in Fig. 2, during the fabrication of release holes, the side length of the square holes expanded from 40 μm to 57.5 μm in $\langle 100 \rangle$ direction and 51.0 μm in $\langle 010 \rangle$ direction, which causes negligible loss to the photosensitive area.

Since the absorption layer was InGaAs layer, the same material as the sacrificial layer, the sidewall of the InGaAs active layer in the release holes need to be protected when the selective etching solution flew to the InGaAs sacrificial layer through the release holes and undergoes etching reaction. Thus, a composite passivation layer was applied on the surface and the sidewall of the devices. According to the reference^{33,34}, Si_3N_4 and SU8 both play an effective role in improving detector performance, silicon nitride (Si_3N_4) can compress dark current promote the detectivity, SU8 can provide better mechanical flexible support. Firstly, a 158nm-thick Si_3N_4 layer was deposited on the top of the device layers by ICP-PECVD and then reactive ion etching was performed to open a $40 \times 40 \mu\text{m}^2$ square release hole in the center of each release hole formed by wet etching above, intervals between platforms and the top contact under the Si_3N_4 layer were also etched out. After that, the devices were coated with 1.15 μm SU8, and then a $26 \times 26 \mu\text{m}^2$ square release hole was opened in the center of each release hole by photolithography, the intervals between platforms and the top contact under the Si_3N_4 layer were also exposed again. As shown in the Fig. 2(b) and (c), the shape of release holes coated with SU8 can be recognized as an inverted trapezoid in $\langle 010 \rangle$ direction and regular trapezoid in $\langle 100 \rangle$ direction. In general, the passivation layer was confirmed to prevent the InGaAs absorption layer from damage during the wet-etching of sacrificial layer.

Finally, Ti/Au top contacts were covered with photoresist, a temporary protective layer to prevent the device layers from being damaged by metal-assisted chemical etching effect³⁵ during the wet-etching of sacrificial layer, and the photoresist layer would be removed after the transfer. After that, the InGaAs sacrificial layer was selectively etched away by immersing the sample in a $\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2:\text{H}_2\text{O}$ (1:8:120) solution, and the metal framed InGaAs PIN detectors detached from the InP host substrate. Subsequently, the flexible membranes were taken out of the etching solution, and slowly dipped into deionized water to remove residual H_2SO_4 and H_2O_2 . Membranes were flipped and attached to a silicon chip coated with photoresist, then n-contact preparation was accomplished by electron beam evaporation. Finally, InGaAs

membranes lifted off from the temporary reusable substrate, and a complete flexible InGaAs PD membrane was prepared, which can be installed in any flexible circuit. In our bending experiment, the flexible InGaAs membrane PD were attached to the indium tin oxide (ITO)/PET flexible sheet (0.05 mm thick, length $L = 35$ mm) with silver conductive adhesive (about 1 μm thick), the whole flexible PD structure was shown in Fig. 1(b). Micrographs of an actual device was shown in Fig. 3(a).

PD Characterization

The flexible InGaAs PIN PD was measured the current-voltage (I-V) in flat state and bent state at different curvature radius(R). The spectral response of the flexible PDs was carried out by a Fourier transform infrared spectrometer at room temperature in a frontside-illuminated format. All current-voltage (I-V) measurements were performed on micromanipulated probe station using a Keithley 4200 semiconductor characterization system at room temperature. An 1550 nm wavelength fiber laser with 2 μm diameter spot was used as a light source for the I-V curves under illumination characteristics. During the bending test, one end of the PET with a flexible PD was fixed and the other end was pushed forward by dL , the bending R is the function of dL ³⁶, as the model shown in Fig. 3(b). The bending tests of devices were performed in two direction: bending up [Fig. 3(c)] and bending down [Fig. 3(d)] for tensile and compressive strains, respectively.

Results And Discussion

Figure 4 shows the spectral response ranges of the InGaAs-based membrane PD (Red line) and the InGaAs PIN rigid PD (Black line, as a counterpart) with the same area fabricated from the same epitaxy wafer. It can be clearly seen that the response wavelength of InGaAs-based membrane PD is the same with that of rigid PD in the range of 640 nm to 1700 nm, which is consistent with other reports^{37,38}. The reason is that the InP contact layer of the flexible InGaAs PIN is 600 nm, and the part of the visible light can pass through the contact layer. According to the previous report, thinner InP contact layer between light source and the InGaAs absorption for the visible-IR InGaAs PD can promote the quantum efficiency in visible light (400 nm – 750 nm) and near-IR (750 nm – 1000 nm) wavelength region^{37–39} and the PD can detect wavelength as short as 400 nm with 200nm-thick InP contact layer, which we will study further and improve our design in the future.

Figure 5(a) and (b) shows the dark current and photocurrent density (J) of the flexible InGaAs membrane PDs in flat condition under various incident optical powers at 1550nm. It can be clearly seen that the PDs exhibit typical rectifying characteristics, indicating an excellent quality of the PIN junction. The dark current density is approximately 2.50 $\mu\text{A}/\text{cm}^2$ at -0.1 V, and there is a gradual increase of the dark current density at the reverse bias region, which could result from the band-to-band tunneling mechanism⁴⁰. A high sensitivity (i.e., the ratio between dark and photocurrent density) of larger than $1.57 \cdot 10^8$ is achieved at 1550 nm. In addition, the PD exhibits a linear response with the excitation power increasing from 20 μW to 275 μW , as shown in Fig. 5(c), indicating that the photogenerated carriers are dominant^{41,42}. The responsivity (R_i) within the range is 0.53 A/W at -0.1 V, corresponding to an external quantum efficiency

(EQE) of 42.46%. The Johnson noise limited detectivity D^* of the device is calculated according to Ref 43, 44:

$$D^* = \frac{q\lambda \cdot EQE}{hc} \left[2qJ + \frac{4k_B T}{RA} \right]^{-\frac{1}{2}} \quad (1)$$

where q , λ , h , c , k_B , T , J , R and A denote electron charge, laser wavelength, the Planck constant, velocity of light, the Boltzmann constant, operating temperature, dark current density, differential resistance and the effective area of the device, respectively, and it can be extracted from the dark current curve that the R_0A of the membrane PD is $2.37 \times 10^5 \Omega \cdot \text{cm}^2$. As a result, the detector exhibits a room temperature D^* of $5.18 \times 10^{11} \text{ cm}^2 \text{Hz}^{1/2}/\text{W}$ @1550nm at -0.1 V, whose performance is the same level with the rigid InGaAs SWIR photodetectors^{45, 46}. Figure 5(d) shows the photocurrent-time response of the flat PDs under a fixed optical power (230 μW), the fall time is about 50.8 μs and the value is a little longer than the previous report, which will be optimized in future.

In order to study the flexible optoelectrical characteristics, the related characterization for the membrane PDs are performed under the bent condition, while the flat condition is referred as the counterpart. Figure 6 shows the PD's dark current density and photoresponse in their flat and bent up/down state. It should be noted that the dark current density increases gradually under bent down condition [Fig. 6(a)] as R reduces. Especially when the R is reduced from 15 mm to 10 mm, the dark current density increases by 5 times. Meanwhile, the photocurrent density of the device is stable when the radius reduced from 30 mm to 15 mm, but decreases by 28% as R is 10 mm. Compared with the bending up condition, the dark current and photocurrent both remain stable under bending down condition, as the radius reduces from 30 mm to 10 mm.

Furthermore, the failure mode of such PDs have been studied deeply. Optical microscope images as shown in Fig. 7 suggest the failure modes of the flexible InGaAs detectors in two bending directions are different. Under bending up condition, cracks are visible at $R = 10$ mm in Fig. 7(a) and (b), the cracks form perpendicular to the forward direction of dL and propagate from one release hole to another one. The cracks lead to the increase of surface leakage current and the decrease of photocurrent. In addition, once the cracking mode occurs, the performance degradation is irreversible. In bending down direction, instead of cracking, we observed that the membrane buckled upward via local delamination from the adhesive at $R = 10$ mm, as shown in Fig. 7(c). The delamination reduces the strain in the membrane thereby preventing the cracking failure mode even at smaller bending radius. Therefore, the device can be bent down to $R = 10$ mm while keeping the integrity and stable photo response of the device. As the bending radius decreases further, the delamination continues to expand, as shown in Fig. 7(d). Even if the bending is released, the membranes do not slip back and stay in delamination forever. Instead of the fracture strains of membranes suffering, the interfacial shear stresses are responsible for the failure behavior and the delamination failure is sensitive to the adhesion strength of film to the substrate³⁶. Hence, increasing the adhesion strength of InGaAs membrane PD to PET helps to overcome delamination.

According to the study about the failure mode of bending silicon ribbons on plastic substrates, the thickness influences the bending mechanics and determines the dominant failure mode, as the thickness increases, the dominant failure mode is as follows: cracking, slipping and delamination^{36, 47, 48}. However, there are two failure modes for the flexible InGaAs PD with a fixed thickness in two bending direction. Unlike the silicon ribbon, the flexible InGaAs PD is a composite multilayer asymstructure, the effect of bending strain on each layer of material is different. As shown in Fig. 1(b), SU8 layer, Si₃N₄ layer and the metal contact coated on the InGaAs PIN membrane structure, asymmetrically from top to bottom. The failure mode is supposed to occur from one layer and transmit to the whole structure. Top and bottom thin metal contact has ductility, and its contribution to the failure membranes can be neglected. The comparison of the Young's modulus for SU8, Si₃N₄ and InGaAs PIN membrane is: SU8 (3–5 GPa) < InGaAs PIN membrane (60–80 GPa) < Si₃N₄ (220–230 GPa). The failure mode is more likely to occur in InGaAs PIN membrane and Si₃N₄. Considering the thickness of the three materials, it can be assumed that, cracking failure dominated in bending up caused by thin Si₃N₄, and the delamination failure dominated in bending down caused by thick InGaAs PIN epitaxy layer. The quantitative analysis of this hypothesis can be further studied, and provide guidance for the bending performance optimization.

In general, the ratio between dark and photocurrent density of the PDs remains stable, larger than $7 \cdot 10^8$ under bending up condition with R decreasing from 30 mm to 15 mm and bending down condition with R decreasing from 30 mm to 10 mm, respectively, indicating that the simplify fabrication of flexible InGaAs PIN membrane PD is feasible.

Fatigue test of the flexible InGaAs-based PD is further performed with a self-assembled slide stable system. The PD is bent on the automated slide rail for 1, 10, 100, and 1000-times bending down cycles, respectively. As shown in Fig. 8, the ratio of photo to dark current density remain more than $4 \cdot 10^8$, while the responsivity remains above 0.49 A/W and thus no failure occur after the bending cycle. These results suggest that such PDs have an excellent durability of the optoelectrical performance after multiple mechanical deformations. Therefore, it could be believed that such gratifying robustness of the flexible InGaAs-based PD with the simplified fabrication process is promising to be applied in the flexible optoelectronic field.

Conclusion

In this paper, we have demonstrated the flexible InGaAs-based membrane PD with the detectivity of $5.18 \times 10^{11} \text{ cm} \cdot \text{Hz}^{1/2} / \text{W}$ by a top-to-down fabrication technique using a sacrificial layer and transferring them onto a flexible host carrier. A side wall passivation with SU8 and Si₃N₄ is applied to simplify InGaAs PD fabrication process. And a large-area flexible PD with operating wavelength from 640 nm to 1700 nm is obtained and the response time is about 50.8 μs. Furthermore, the results reveal that the over-all optoelectrical performance of such flexible PDs keeps stable in bending state. In addition, fatigue test suggests that cracking and delamination modes dominates in bending up and down conditions, respectively. We believe that the demonstrated flexible III-V material-based PD with the high performance

of the rigid PD is step forward in merging mature compound semiconductor technology and flexible optoelectronic device with simple design and high performance for broad spectrum detection.

Declarations

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Author Contributions

X.L. and Z.D. conceived the idea. X.L. conducted the device fabrication. J.Z., C.Y., X.T. and Z.G. equally to this work for assisting the sample preparation and characterization. H.C. supervised the entire research project. The manuscript was written by X.L. and Z.D. All authors have reviewed the manuscript and given their approval to the final version of this manuscript.

Additional Information

Competing financial interests: The authors declare no competing financial interests.

Data availability

All data generated or analysed during this study are included in this published article (and its Supplementary Information files).

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Figures

Figure 1

Device fabrication: (a) A flow chart for fabrication process: (i) Material growth of InGaAs PIN PD, (ii) Deposition of Up-contact metal frame. (iii) Preparation of release holes array, (iv) Preparation of passivation layer, (v) Preparation of electrode protective layer, (vi) Peeling off the InGaAs PIN membrane PD, (vii) Deposition of bottom contact and transfer of devices. (b) Cross-sectional structure of a fabricated InGaAs PIN PD based on ITO/PET flexible substrate.

Figure 2

SEM image of etch holes in fabricated InGaAs PD based on the InP substrate: (a) The surface of the etch holes array. (b,c) The cross section of side wall covered with composite passivation layer in [100] and [010] direction. (d) A magnified view of a side wall of the outline area on the left shows side wall covered with Si_3N_4 (158 nm) and SU8 (1.15 μm).

Figure 3

(a) Zoom-in views of a fabricated large area (3 × 3 mm²) InGaAs PD on flexible PET substrate. (b) Schematic diagram for the measurement principle and the corresponding model. (c, d) A schematic diagram of fabricated flexible InGaAs PD under bending up and bending down test, respectively. The bottom left insets provide schematic illustrations of the bending geometries.

Figure 4

The response Spectrum of InGaAs Flexible membrane PD and InGaAs rigid PD at 300K

Figure 5

J-V responses of the InGaAs flexible detector under flat condition: (a) At darkness and (b) Different input optical power levels. (c) Photocurrent (PC) as a function of the optical power, the solid line represents a linear fitting of the experimental data. (d) Dynamic photocurrent behavior.

Figure 6

Device characterization under bent conditions: J-V responses of the detector under bending up and down condition. At darkness: (a,c) and Illumination: (b,d) at 1550 nm ($P=230 \mu\text{W}$).

Figure 7

Microscope image of bending flexible InGaAs detector: (a) Bending up of $R = 10$ mm. (b) A magnified view of the outline area on the left shows the cracking failure surface. (c) Bending down of $R = 10$ mm shows the appearance of the delamination failure. (d) Bending up of $R < 10$ mm shows the expansion of the delamination failure.

Figure 8

Devices characterization after multiple bending down cycle of 1, 10, 100, 1000 times (a radius of $R = 15$ mm): Left: Dark and photocurrent (PC) density of the devices. Right: Responsivity of the device at 1550 nm ($P = 230 \mu W$).

Supplementary Files

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- [SCSupplement.docx](#)