Supplementary Materials for

**Breaking the absorption limit of Si toward SWIR wavelength range via strain engineering**

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The PDF file includes:

- Supplementary Text
- Legends for movies S1 and S2
- Figs. S1 to S18

Other Supplementary Material for this manuscript includes the following:

(available at advances.sciencemag.org/cgi/content/full/6/31/eabb0576/DC1)

- Movies S1 and S2
Supplementary Text
Fracture Trend of Si NMs of Different Thickness Under Increasing Straining Pressure

The theoretical elastic limit of the perfect bulk Si crystal is ~17% (28); however, the maximum applied strain in large-area Si samples achieved via the most commonly adopted strain engineering approaches (use of a SiN stressor, lattice mismatch epitaxy, and oxidation-induced residual strain by releasing a host Si substrate of a SOI wafer) was reported to only be approximately 1% (16, 23, 25). The maximum applied strain in the aforementioned approaches was restricted by the limited lattice-constant difference in epitaxy methods (23, 25) and a maximum possible residual stress of ~6 GPa achieved via volume expansion during SiO₂ growth in the oxidation-induced approach (16). In our straining approach, we can mechanically stretch the Si NMs until the lattice fractures, and thus, we can apply a much higher strain. To achieve the maximum applied strain in PD pixels, the optimizations of thickness and size of the Si NM-based MSM device is important. Therefore, prior to the fabrication of Si NM array-based PD devices, we investigated the effect of the thickness and size of Si NM on the applied strain to realize the maximum biaxial strain. The square-shaped Si NMs (20 × 20 µm²), having different thicknesses of 50, 20, 10, 8, and 4 nm, were bonded on PI films and subsequently mounted on the bulge test setup for pressure-strain calibration. The strain values obtained from Raman measurement shown in Fig. 2B indicate that the maximum applied biaxial strain value gradually increases up to a thickness of 10 nm (4.36%), and thereafter decreases to a relatively lower value (4.1% for a 4-nm thick Si NM).

To understand this anomalous strain reduction trend beyond a particular thickness, we continuously recorded the optical images for each thickness during Raman measurement under increasing pressure, and the images are presented in the fig. S3A. It can be noticed that the cracks in the Si NMs with thickness exceeding 10 nm were aligned at an angle of 90° under the influence of gas pressure-induced biaxial tension, whereas for the samples with thickness below 10 nm, the cracks were oriented randomly. The Si NMs with thickness exceeding 50 nm did not crack even until the PI film blown up under a very high pressure. Because the Si NMs were firmly bonded on the PI film, the increasing pressure resulted in the simultaneous expansion of the Si NM and the PI film. The expansion trend in the top Si NMs was strongly influenced by the expansion in the underneath PI film. For sufficiently thick Si NMs (≥ 10 nm), the inherent mechanical and structural properties of the top Si NMs predominantly govern the expansion. For ultrathin Si NMs (with thickness less than 10 nm), the fracture initiated at the defective lattice sites, and it subsequently propagated across the entire nanomembrane at a higher applied pressure. Because the Si NMs used in this study were of <100> orientation, the fracture occurred in the <010> and <001> planes of the Si lattice under uniform biaxial tension, thereby leading to crack propagation at an angle of 90°. In contrast, in the thin Si (≤ 8 nm), having less mechanical strength and a relatively large number of lattice defects at the edges and on the top and bottom surfaces, the local non-uniform expansion in the underneath
PI provided random fractures at a relatively low applied pressure than that in the thick NMs, thereby resulting in the saturation and reduction of the maximum applied strain value. Once the first crack originated at a particular strain under a sufficient pressure, without any further increase in pressure, the number of cracks was increased within a couple of seconds (fig. S3B). In the 10-nm thick Si NM sample, the first crack initiated at approximately 4.36% strain, and thereafter, the number of cracks increased immediately at the same pressure (fig. S3B (i–vi)). Further increase in cavity pressure resulted in an increase in the number of cracks instead of the strain value. The strain energy in the Si lattice was released with the generation of each new crack under increasing pressure (31), which maintained the strain at the particular value applied at the origin of the first crack. It can be noticed that the cracks in the Si NMs with thickness exceeding 10 nm were aligned at an angle of 90⁰ (Inset of Fig. 2A) whereas for the samples with thickness below 10 nm, the cracks were oriented randomly (fig. S3A and S3B). Random fractures at a relatively low applied pressure resulting in the saturation and reduction of the maximum applied strain value.

**Computational Details**

Calculations are performed within the framework of density functional theory, as implemented in the Vienna ab-initio simulation package (VASP) code (32). The ion-electron interaction is described using the projector-augmented wave (PAW) method (33), The exchange-correlation potential is treated in the generalized gradient approximation (GGA) (34). An energy cutoff of 500 eV is adopted for the plane wave expansion and a 16×16×1 Monkhorst–Pack k-point grid is used for an integration over the two-dimensional (2D) Brillouin zone. In this calculation, the in-plane lattice constant, 5.43 Å, is adopted and a vacuum region is accounted for more than 15 Å. All of the atomic positions were fully optimized using the conjugate gradient method for the geometry optimization. To consider the same thickness of the experimental sample, here we construct an ultrathin Si nanomembrane (NM) model with thicknesses of ~10.7 nm (80 Si layers) as shown in fig. S4A. In addition, the dangling bonds on both top and bottom surfaces for Si NM are passivated with oxygen atoms. Because the ultrathin Si NM can be viewed as 2D-like structures, the surface Brillouin zone (BZ) instead of the bulk Brillouin zone was used for the calculation as shown in fig. S4A.

**I–V Characteristics and Photoresponse of a Single PD Pixel MSM and p-n Junction Devices**

The devices designed with MSM pixels usually have slow and relatively modest photoresponse in comparison to the devices fabricated with p-n or p-i-n configurations. To compare the optoelectronic performance of MSM and p-n pixels fabricated with low dimensional Si, we also fabricated p-n diode devices on 100- and 10- nm thick Si nanomembrane via selectively implanting Arsenic ions. The p-n junction pixels fabrication steps are schematically shown in fig. S5A, whereas fig. S5B shows the optical images of ion implanted regions of SOI wafers and fabricated devices on 10 and 100 nm thick Si nanomembranes. The I-V characteristics of p-n pixels fabricated on Si NM of different thickness are presented in fig. S6. The p-n pixel fabricated on 100 nm thick Si clearly shows a rectification behavior with clean dark current. On the other hand, the I-V recorded under dark for p-n pixel on 10 nm thick Si NM reveals a symmetric characteristic with negligible rectification. The current-voltage characteristics (recorded under dark and illumination) of p-n pixel fabricated on 10 nm thick Si NM shows very similar features to that of MSM device of same thickness. The current–voltage (I–V) characteristics of a single MSM PD pixel recorded in the dark and with increasing incident optical powers of 520-nm light (fig. S8A) exhibited an increase in photocurrent with increase in the light intensity and the applied bias. The obtained linear current–voltage characteristics for ±1 V signify a typical MSM photosensing feature under the photoconductor mode provided by ohmic contacts between Cr/Au and the Si NM. A low doping level and a reduced Si NM thickness of 10 nm resulted in a lower current value in the dark condition (in the order
of $10^{-10}$ A) and a noisy I–V nature (fig. S8B) possibly because of the presence of an unavoidable thin native oxide at the Si surface.

The electronic transport in ultrathin Si NMs is sensitive to the surface conditions and strongly influenced by surface trap states because of the high surface-to-volume ratio. The sheet density of dopants in thin membranes ($10^{-9} \text{ cm}^{-2}$ for a nominal doping level of $10^{15} \text{ cm}^{-3}$) is typically lower than the surface trap density ($10^{-11} \text{ cm}^{-2}$) for ultrathin Si (~10 nm) containing native oxide (35). Therefore, a moderately doped 10-nm thick Si NM exhibited a highly resistive nature with noisy I–V characteristics. For the conventional photodetection and photovoltaic applications, the vertical p–n or p–i–n device architectures are preferred and adopted on the bulk Si platform. The strain-based investigations conducted on ultrathin Si NMs in this study restricted the implementation of vertical p–n or p–i–n structures. The fabrication of such junctions is possible in lateral geometry; however, the electron transport in ultrathin Si NMs is highly sensitive to the surface and is governed by the surface characteristics irrespective of the dopant type in the inner Si core (as long as the doping is not degenerate) (35). Therefore, having several fabrication complexities, the lateral p–n or p–i–n junctions can provide I–V characteristics similar to that of the MSM structure and not very favorable over the MSM structure.

To qualitatively quantify the light absorption in Si NMs of different thickness, we have carried out the photocurrent measurement on Si NM PD pixels fabricated on 20-, 10- and 4-nm thick Si NMs. Under a fixed exposure of 532 nm light, the PD pixel fabricated at 20-, 10- and 4-nm thick Si NM exhibited a photocurrent of $6.20 \times 10^{-7}$, $4.29 \times 10^{-8}$ and $4.29 \times 10^{-9}$, respectively (fig. S9). The photocurrent value recorded under the fixed light exposure found decreasing with decrease in nanomembrane thickness because of the reduced optical absorption in thin nanomembranes. A comparison of photoresponse obtained from 20 and 10 nm thick Si nanomembrane under increasing incident optical power and increasing strain is shown in fig. S10. The responsivity values and effect of strain on thick Si nanomembrane sample is found to be more pronounced. However, the thick sample possesses lower value of biaxial strain, therefore lower thickness is preferred for broad wavelength tunability. In the practical applications of photodetectors such as image sensor, a constant responsivity over a wide range of incident light intensity with sensing capability for very weak light is critically important. The linear dynamic range (LDR) is a crucial parameter for photodetectors which characterize the incident light intensity range in which the photodetectors have a constant photo response. The LDR is obtained by $\text{LDR} = 20\log \left( \frac{P_{\text{max}}}{P_{\text{min}}} \right)$, where $P_{\text{max}}$ is the maximum intensity beyond that photocurrent begins to deviate from linearity and $P_{\text{min}}$ is the minimum detectable light intensity and represents the noise equivalent power (NEP). To estimate the LDR of our fabricated device, we carried out the photocurrent measurement on the MSM PD pixel fabricated on 10 nm thick Si NM under the exposure of the 532- and 980-nm light with varying intensity (fig. S11). The photocurrent was increased linearly with increasing irradiance up to $\sim 105.06 \text{ mW/cm}^2$ and $1.02 \text{ mW/cm}^2$ for the incident light of 532- and 980-nm, respectively. The fabricated MSM PD pixel exhibited nearly similar minimum detectable light intensity of 0.2 mW/cm$^2$ for both wavelengths. These values of $P_{\text{max}}$ and $P_{\text{min}}$ resulted in the LDRs of 67.6 dB and 25.8 dB for 532 and 980 nm light, respectively. The obtained LDR values of the fabricated MSM PD pixel in this work are slightly lower than that of the previously reported which is mainly attributed to the higher NEP originated due to the noisy dark current (38), which restricts the clear distinguish between dark and phot current under low incident intensities. In addition, the obtained LDR value for 980 nm light is low because our instrument has a limited light intensity of 1.02 mW/cm$^2$ for this wavelength.

Applying Strain into Si Nanomembrane Using Textured SiO$_2$/Si Substrate

The optical band-gap near the band-edge and its tunability with strain can be determined using UV-Vis or photocurrent spectroscopy. Most of the UV-Vis spectrometers are closed systems which work in visible wavelength range under transmittance mode. Coupling a bulge test set-up with all the gas pipelines inside
the UV-Vis system is challenging. Additionally, we need to record the Raman spectra at each step of increasing gas pressure to measure the applied strain in the nanomembrane pixels, for that we need a Raman system coupled with the UV-Vis spectrometer. Similarly, to record external quantum efficiency (EQE) under different strain values, we need a quantum efficiency measurement system coupled with Raman system which is also a critically challenging task. Therefore, to investigate the effect of strain on the band edge of Si, we adopted another straining approach reported in literature instead of bulge testing method \((39, 40)\). In this approach, we transferred 10 nm thick Si nanomembrane samples on a SiO\(_2\)/Si substrate containing nanotexture arrays. The linear strips having sharp top edges were used for uniaxial strain, whereas conical shaped textures were used for applying biaxial strain (fig. S12). This straining approach provides relatively low values of strain in comparison to the bulge testing; however, it provided the opportunity to fabricate complete MSM devices on SiO\(_2\)/Si substrate, thus facilitated the measurement of photocurrent near band edge. In order to apply uniaxial and biaxial strain into Si NMs, we first texturized \(\sim 1 \times 1\) cm\(^2\) bulk Si wafer into strips and cones via standard lithography and RIE processing as shown in the AFM topographic images of (fig. S13A). Thereafter the texturized Si substrates were heated in a tube furnace at 1000 °C in ambient environment for 12 hours. The top surface of Si was exposed to air at elevated temperature and subsequently oxidized into SiO\(_2\), leading to the growth of \(\sim 300\) nm thick SiO\(_2\) layer. Finally, the patterned Si nanomembrane samples were transferred onto the textured SiO\(_2\)/Si substrates from SOI wafers via wet transferring approach. The mechanical deformation of \(\sim 10\) nm thick Si NM through conformal contact with textured SiO\(_2\)/Si substrate during solvent evaporation process resulted in uniaxial and biaxial strain into Si NMs transferred on stripped and cone textures, respectively (fig. S13B and Fig. S13C). The height, separation and tip dimension of textures were optimized to achieve the maximum strain. The applied strain was characterized by Raman spectroscopy (fig. S13D). Si NM transferred on strips exhibited a uniaxial strain of \(\sim 1.36\)%, whereas, Si NM transferred on conical textures showed strain of \(\sim 1.4\)% The strain across the Si NM area was distributed non-uniformly exhibiting maximum value at the tip of texture for both the strip and cone case.

The photocurrent recorded from the tip and base region of the uniaxially strained Si clearly reveals the strain effect via exhibiting higher value of photocurrent when light is exposed at the tip region (fig. S14A). To investigate the effect of strain at optical absorption, particularly near band gap, we recorded photocurrent for both the uniaxially and biaxially strained Si NM devices using tunable light exposure originated from a supercontinuum laser source coupled with a monochromator. The photocurrent was recorded with a combination of a lock-in amplifier and optical chopper used to detect the ultralow photocurrent signals. fig. S14B displays the recorded photocurrent spectra for unstrained, uniaxially and biaxially strained Si NM PD pixels fabricated on planar, strip and conical textured SiO\(_2\)/Si surfaces. The unstrained Si shows the photocurrent onset at the indirect band gap edge of around 1.1 eV, whereas an extension in the photocurrent onset can be clearly noticed for both the uniaxially and biaxially strained Si samples. The strain-induced band shrinkage shifts the cutoff toward longer wavelengths. The biaxially strained Si sample exhibits a more pronounced strain effect with the absorption onset up to a wavelength of 1170 nm which is in close agreement with the calculated reduction of the band gap at this level of applied strain.

Integration of PbS quantum dots onto Si NM surface

The obtained photoresponsivity in our strained Si NM PD pixels is relatively low for SWIR range (\(~35\) and \(4 \mu\)A/W for wavelengths 1310 and 1550 nm, respectively) in comparison to that obtained in visible wavelength range. We tried to address this issue via integrating PbS quantum dots (QDs) onto the Si NM surface. The quantum dot sensitization is a well investigated approach to enhance the optical absorption and extension of wavelength range with maintaining the flexibility and stretchability of low dimensional systems \((41)\). In the view of SWIR optical sensing, we adopted the PbS QDs, which possesses narrow
band gap and large excitonic Bohr radius enabling a strong quantum confinement effect for strong optical absorption in SWIR wavelength range. To sensitize Si NM with quantum dots, a homogeneous dispersion of PbS QDs ($\lambda_{\text{Emission}} = 1600$ nm) in toluene (10 mg/ml) was spin coated on 10 nm thick Si NM MSM PD pixels and dried overnight in vacuum oven at 70 °C before optical characterization. fig. S16A shows the schematic representation of the device structure along with the optical images of bare and Si NM PD pixels integrated with PbS QDs. The photocurrent spectra recorded for the bare and QDs sensitized unstrained Si NM devices under the modulated exposure of 1550 nm light of 50 mW is shown in the fig. S16B. The Si NM device decorated with PbS QDs exhibited a clear photoresponse under the 1550 nm light exposure. On the other hand, bare unstrained device did not show any change in photocurrent. The QD sensitized device exhibited a photocurrent of $\sim 1 \times 10^{-8}$ with an optical responsivity of $\sim 12.5$ mA/W, which was comparable to that obtained for visible light of similar intensity. The obtained results clearly reveal that the PbS QD sensitized MSM PD pixel devices fabricated on ultra-thin (~10 nm thick) Si NM could be successfully employed for SWIR imaging application with reasonable optical responsivity. It may be noticed that our main interest was to achieve the SWIR optical activity in intrinsic Si without any alloying, doping or surface modifications, which is very crucial for the integration of such SWIR image sensors under CMOS platform. That motivated us to focus on the strain induced band gap modification in pristine Si and extending the absorption onset, therefore we carried out prototype device fabrication with bare Si NMs.

Applying Strain into Si Nanomembrane Using Radial Tensioning Stage

Under the gas pressure-induced biaxial tension, the area at the topmost part of the hemisphere tends to stretch more in comparison with the area near the periphery, which results in an increase in strain in the inner pixels of the PD matrix. To achieve a uniform strain in each PD pixel, we attempted to stretch the PI film containing the Si NM pixels horizontally using a specially designed radial tensioning stage. The designed tensioning stage contains eight circular metal plates that tightly hold the PI film and apply uniform mechanical biaxial tension on the bonded Si NM pixels (fig. S18A). The screw-based scrolling mechanism attached to each metal plate provides a controlled back and forth movement in the radial direction. The PI film containing the Si-NM matrix array was firmly bonded to each of the eight metal plates of the tensioning stage using screws. To apply strain on the Si NM pixels, each metal plate sequentially stretched outwards in intervals of $\sim 200$ µm using rotating screws, enabling biaxial stretching in the mounted PI film. Once the stretching step was completed for all the eight plates, the next round followed. The applied strains on the Si NM pixels were measured at the end of each stretching cycle. In this manner, we stretched the Si NMs until they fractured, and the applied maximum strain was measured. fig. S18B presents the measured strains on the Si NM pixels; it indicates that the strain distribution across the 10-nm thick Si NM pixels was non uniform, with maximum and minimum strains of 2.86% and 1.98%, respectively. The maximum applied strain using tensioning stage is relatively less than that obtained using the gas pressure-induced bulging approach.

movie S1. Demonstration of the imaging capability under SWIR light.

Movie clip shows the real time imaging of “Y” shape 1310 nm light projected onto Si NM PD array system subjected to different strain levels. Left panel shows the real-time photocurrent map in a normalized color-coded scale generated using the output data collected from each PD pixel via DAQ unit. Right panel shows the measurement system consisting PD array device mounted on bulge test setup, a fiber guided 1310 nm laser light, an IR card to visualize the incident laser pulse. It can be clearly noticed that there is
no photoresponse from the PD pixels, when they are at zero strain level. However, as the light is exposed onto the PD array subjected to a maximum biaxial strain of ~3.5%, a clear on-off representing “Y” shape can be realized.

**movie S2. Imaging with concave geometry.**
Movie clip shows the real time imaging of 532 nm light beam focused with a plano-convex lens on Si NM PD matrix array under flat and concave geometry. Left panel shows the real time photocurrent map in color coded scale (inset schematically shows the corresponding curvature of PD surface). Top right panel shows the video of Si NM PD pixels under flat and bulged condition making PD pixel surface to concave geometry. Bottom right panel demonstrate the overall measurement system exposed with modulated laser beam coupled with plano-convex lens. When the PD pixels are arranged in flat condition, a non-uniform photocurrent distribution with lower photocurrent in inner pixels can be clearly noticed. Whereas, under the influence of straining pressure the PD pixels arranged in concave geometry provides a nearly uniform photocurrent pattern across the PD matrix with increased photocurrent in the inner pixels.
**fig. S1.** Photographs of transferring process and mounted 6×6 Si NM photodetector array on bulge test set up. Photo Credit: Ajit K. Katiyar, Yonsei University.
**fig. S2.** Schematic diagram showing the details of used bulge test set up used for applying strain in Si nanomembranes.
fig. S3. **Optical images of the Si NM under straining pressure.** (A) Optical images of Si nanomembranes before and after applying strain, (B) Fracture generation in 10 nm thick Si NM. After first fracture the number of cracks increase with increase in pressure leading to maintain the strain in membrane (i-vi). The fracture at an angle of 90° up to a thickness of 10 nm shows the fracture from typical (100) planes.
fig. S4. Schematic atomic arrangement of top Si and Brillouin zone. (A) Top view of 10.7 nm thick Si nanomembrane (NM) model, (B) Bulk (bottom) and surface (top) Brillouin zone (BZ).

fig. S5. Fabrication of p-n PD pixels. (A) Schematic representation of the fabrication of p-n junction pixel via ion implantation. (B) Optical images of the ion implanted region on SOI wafer and the fabricated p-n junction PD Pixels on 10 and 100 nm thick Si nanomembrane.
**fig. S6.** I-V characteristics of the p-n junction diodes recorded under dark and under light exposure. (A) Fabricated on 100 nm thick Si nanomembrane (B) Fabricated on 10 nm thick Si nanomembrane.

**fig. S7.** Optical image of Si NM photodetector array mounted on bulge test set up during measurement of photoresponse under 532 nm light with applied straining pressure. Photo Credit: Ajit K. Katiyar, Yonsei University.
fig. S8. IV characteristics of 10 nm thick Si NM device under 520 nm light. (A) with increasing incident optical power, (B) Semi-log IV characteristics with increasing strain.

fig. S9. Photocurrent characteristics of the MSM PD pixel devices fabricated on 20-, 10- and 4- nm thick Si nanomembranes.
fig. S10. Comparison of photoresponse of 20 and 10 nm thick Si nanomembrane. (A) with increasing incident optical power. (B) with increasing strain.

fig. S11. Photocurrent measured at −1 V under the exposure of different optical power to extract the linear dynamic range (LDR) of the Si NM PD pixel for (A) 520 nm (B) and 980 nm light.
**fig. S12.** Schematic representation for the fabrication of strained Si NM PD pixel via transferring Si nanomembrane on textured SiO$_2$/Si substrates.

**fig. S13.** Structural characterization of strained Si NM PD pixel fabricated using textured substrates. (A) AFM topographic images of the prepared strip and nano-cone textured SiO$_2$/Si substrates. (B) Optical and (C) SEM images of fabricated devices on the Si NM transferred on strip and nano-cone textured surface for the application of uniaxial and biaxial strain. (D) Raman spectra of the uniaxially and biaxially strained Si nanomembranes.
fig. S14. Photosensing characteristics of 10 nm thick Si NM transferred on textured substrate. (A) I-V characteristics recorded for uniaxially strained Si NM under the exposure of the laser at strained and unstrained region. Inset shows the optical photograph of the laser spot exposed at different position. (B) Photocurrent spectra measured for the unstrained, uniaxially strained and biaxially strained Si NM samples. The extension on the photocurrent onset for biaxially strained Si can be clearly noticed.

fig. S15. Wavelength dependent responsivity measured at different strain level. Plots clearly shows the photo sensing capability of 10 nm thick Si NM device beyond Si photo absorption wavelength range (400-1100 nm) under applied strain. Device can detect 1310 nm light above ~1.8% and 1550 nm light above ~3.2% applied bi-axial strain.
fig. S16. Integration of PbS quantum dot onto Si NM PD pixels. (A) Schematic representation of Si NM PD pixel devices integrated with PbS QDs and corresponding optical images of bare and QD sensitized Si NM device. (B) Photocurrent characteristics of bare and PbS QD sensitized MSM PD pixel devices recorded under the modulated exposure of 1550 nm light.

fig. S17. Strain distribution profile of 6×6 Si NM array with an applied pressure of 1600 Torr.
**fig. S18. Photographic image of planar stretching machine and obtained strain profile.** (A) Photographic images of planar stretching machine to apply strain into Si NM pixels. Photo Credit: Ajit K. Katiyar, Yonsei University. (B) measured strain into Si NM PD pixels stretched via radial tensioning machine up to the fracture into Si nanomembrane.