

Wrinkled Single-Crystalline Germanium Nanomembranes for Stretchable Photodetectors

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Abstract—Germanium nanomembranes are suitable for flexible electronics, including high-mobility nonsilicon transistors, fast radio-frequency switches, microwave diodes, and high-performance photodetectors. In order to enhance the flexibility of the germanium-based devices, we present a strategy to integrate single-crystalline germanium nanomembranes into a wave-like wrinkled geometry with a uniform periodicity and amplitude on elastomeric substrates. Wrinkled single-crystalline germanium nanomembranes are realized with a reversible and large deformation up to 10%, and the stretchable metal–germanium–metal photodetectors have been demonstrated. Optoelectronic response studies reveal that the wrinkled germanium-based photodetectors exhibit enhanced efficiency of optoelectronic interactions compared with planar photodetectors using flat germanium nanomembranes. Furthermore, the wrinkled photodetectors reveal high response speed and stretchable capability of up to 8.56%. This paper may pave the way for the integration of germanium nanomembranes into the field of flexible/wearable optoelectronics.

Index Terms—Germanium nanomembranes, light absorption, photodetectors, stretchable electronics, wrinkles.

I. INTRODUCTION

HIGH-PERFORMANCE flexible and stretchable electronics are demanded by paper-like devices [1], integrated

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circuits [2], photovoltaic solar cells [3], and biointegrated devices [4]–[10] due to the unique features, such as large area coverage, good tolerance of mechanical deformation, wearable properties, lightweight, and biocompatibility. The materials can be produced by integrating single-crystal semiconducting nanomembranes, such as Si [11], Ge [12], GaAs [13], and GaN [14] into elastomeric substrates like polydimethylsiloxane (PDMS). Owing to the high electron/hole mobility [15], low-field drift velocity [16], small bandgap, and superior mechanical properties [17], nanoscale germanium membranes are desirable in flexible transistors [18], radio-frequency diodes [19], [20], and photodetectors [21], [22]. These flexible devices demand that the germanium nanomembranes are not only bendable, but also stretchable and tolerable to large deformation [23] in a conformal manner [24]. However, present flexible devices based on germanium nanomembranes can only be bent but not stretched [12], [16]–[22], thereby hampering wider application of flexible germanium-based devices.

Semiconducting nanomembranes with the proper strain engineering can be rearranged to form wrinkles or buckles with uniform “wave” geometries on elastomeric substrates [25]–[28] and these wrinkles or buckles can accommodate vast compressive or tensile strain [13], [29], [30], and thus paving the way for their use in stretchable or curvilinear devices. Here, wrinkles are introduced into single-crystalline germanium nanomembranes to build their stretchable capability, and the wavy germanium nanomembranes are further utilized to produce the stretchable photodetectors. The wrinkled germanium nanomembranes can tolerate large deformation of up to 10% with excellent reversibility and the photodetector shows fantastic optoelectronic response in conjunction with highly stretchable capability compared with current materials. This paper may provide a strategy for the applications of germanium nanomembranes in the flexible and stretchable optoelectronics.

II. EXPERIMENT

A. Fabrication of Wrinkled Germanium Nanomembranes

Generation of germanium wrinkles on the elastomeric substrate such as PDMS is schematically shown in Fig. 1. The patterned n-type (phosphorus-doped) germanium nanomembrane with a thickness of 50 nm was released from the germanium-on-insulator (GOI) wafer by photolithography, reactive ion etching (RIE), and selective wet chemical etching. The GOI wafer consists of 50-nm top Ge nanomembrane, 200-nm buried SiO₂ layer, and silicon substrate. A photoresist layer

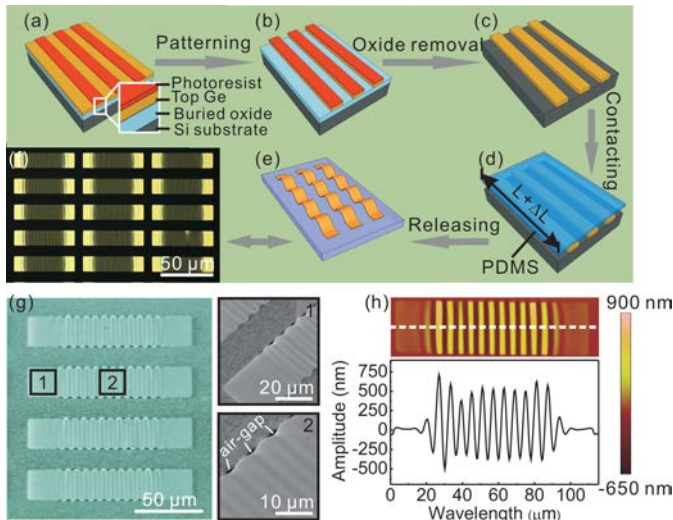


Fig. 1. Fabrication and morphologies of germanium nanomembrane wrinkles. (a)–(e) Schematic of the process to fabricate the strain-engineered wrinkles in the germanium nanomembrane. (f) Optical microscopic image of the typical germanium wrinkle array. (g) Tilted view of germanium nanomembrane wrinkle by SEM (left), as well as the morphologies of the wrinkle at the edge (marked as “1”) and middle region (marked as “2”). Air gaps between germanium wrinkles and PDMS substrates are highlighted. (h) AFM image of the wrinkles (top) and corresponding surface profiles (bottom).

(AZ 5214) was spin-coated on the GOI wafer and heated to 100 °C for 3 min. The rectangular patterns (30 $\mu\text{m} \times 120 \mu\text{m}$) were formed by UV exposure through a photomask [Fig. 1(a)] and the patterns were transferred to the germanium layer of the GOI by RIE [Fig. 1(b)]. The patterned GOI wafer was immersed in HF (40%) to remove the underlying buried oxide layer to release the germanium nanomembrane, which was subsequently adhered to the silicon substrate [Fig. 1(c)] because of the van der Waals force between the released germanium nanomembrane and silicon substrate [31]. Therefore, the pattern alignment defined by the lithography process could be preserved. A prestressed PDMS with a strain value ε_{pre} defined by $\Delta L/L$ was put close to germanium nanomembrane to form intimate contact [Fig. 1(d)]. After peeling the prestressed PDMS off from the silicon substrate, germanium nanomembranes were detached and transferred to the prestressed PDMS. Release of the prestrain in the PDMS resulted in the formation of the wrinkled germanium nanomembrane arrays, which possess better flexibility compared with flat counterpart, as shown in Fig. 1(e) (sketched) and Fig. 1(f) (optical microscopic image).

B. Fabrication and Characterization of Flexible Photodetectors

The as-fabricated germanium nanomembrane wrinkles on PDMS substrate were fixed on a homemade fixture which can stretch the wrinkles with different strain values by tuning the distance between the two anchor sides of the fixture. To exhibit the flexibility of the germanium wrinkles in forms of functional electronic devices, metal–semiconductor–metal photodetectors (MSM PDs) were constructed with the as-fabricated germanium wrinkles. We constructed MSM PDs with germanium wrinkles on PDMS by magnetron sputtering platinum (Pt $\sim 100 \text{ nm}$) to form Schottky electrodes through

a shadow mask without postannealing treatment, and a typical optical microscopic image is shown in the bottom left of Fig. 3(a). The electrical properties and the optoelectronic responses were explored with a semiconductor parameter analyzer (Keithley 4200) at room temperature, and the light source was provided by a normal white fluorescent lamp with tunable light densities (from 0 to 25 $\text{mW} \cdot \text{cm}^{-2}$).

III. RESULTS AND DISCUSSION

A. Characterization of Germanium Nanomembrane Wrinkles

The morphology of the germanium nanomembrane wrinkles are characterized by scanning electron microscopy (SEM) and atomic force microscopy (AFM) as shown in Fig. 1(g) and (h). Fig. 1(g) (left) reveals a tilted view of the morphology of germanium wrinkle and the prestrain applied to the PDMS stamp is about 10% by elastic stretching. The results indicate that the germanium nanomembrane has superior mechanical flexibility that can be extruded into a regular wave-like structure with a regular periodicity. The magnified morphology of the wrinkles at the edge and the middle areas marked by black frames are shown in Fig. 1(g) (right). The edge area (region 1) of the wrinkles has a flat and stable configuration resulting from the traction-free edge [32] and air gaps can be observed from the interface between PDMS and germanium nanomembrane in the middle part (region 2). Fig. 1(h) (top) shows the 3-D profile of the wrinkles examined by AFM. To examine the periodicity and amplitude of the germanium wrinkles, a line scan is obtained from the AFM image along the direction perpendicular to the wrinkle [Fig. 1(h) (bottom)]. The wrinkle is sinusoidal with a periodicity (λ) of 6.12 μm and the amplitude (A) is about 460 nm. The wrinkling process can be well understood by the finite deformation theory with the wavelength (λ) and amplitude (A) [33]

$$\lambda = \frac{\lambda_0}{(1 + \varepsilon_{\text{pre}})(1 + \zeta)^{1/3}} \quad (1)$$

$$A = \frac{A_0}{\sqrt{1 + \varepsilon_{\text{pre}}(1 + \zeta)^{1/3}}} \quad (2)$$

where ε_{pre} is the prestrain, λ_0 and A_0 denote the wavelength and amplitude of the initial buckled geometry based on nonlinear analysis [29], and ζ can be written as $\zeta = 5\varepsilon_{\text{pre}}(1 + \varepsilon_{\text{pre}})/32$. According to (1) and (2), the theoretical wavelength and the amplitude are 7.11 μm and 749.9 nm, respectively. The slight disparity between the theoretical and experimental results can be attributed to the partial delamination of the films from the substrates as suggested by SEM images [Fig. 1(g)], and finite size effects considered for the films [29].

B. Stretchable Capability of Germanium Wrinkled Nanomembranes

Before the functionalization of these germanium nanomembrane-based 3-D wrinkles, the corresponding stretchable properties are needed to be investigated. In this respect, we first investigate the evolution and stretchability of the wrinkles in detail, and the strain applied to the PDMS

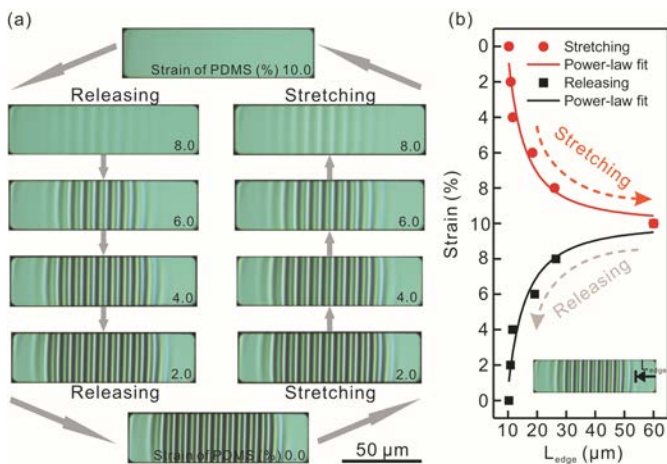


Fig. 2. Stretchability of germanium wrinkles. (a) Morphology evolutions of germanium wrinkles as the strain of PDMS substrate altering. (b) Length of the flat edge in germanium wrinkles varies with the strain of PDMS during releasing (black square dots) and stretching (red circular dots) process. Both experimental results are fitted by power-law function.

is manipulated to cause either extrusion or stretching of the germanium wrinkles. The morphological changes during the deformation of the PDMS substrate are monitored by optical microscopy as shown in Fig. 2(a). The germanium nanomembrane is transferred to a prestrained PDMS substrate with the prestrain of 10%. When the prestrain is released to 8%, small wrinkles start to form in the middle of germanium nanomembrane. The wrinkles propagate toward the edges symmetrically with increasing amplitude as the prestrain is gradually reduced. The origin and propagation of the wrinkles in the germanium nanomembrane can be modeled by dynamic mechanics in which the interface interaction is considered [34]. When the prestrain is totally released, the wrinkles have a stable and wavy configuration with the maximum amplitude as shown in the bottom image in Fig. 2(a) (marked as 0.0). It should be noted that the strain is generated in wrinkled germanium nanomembrane as the prestrained PDMS is released, while the value is much lower than that in the prestrained PDMS. This is critically important to integrate germanium wrinkles into the applications in stretchable/flexible electronics, since the external strain can be highly weakened in the wrinkled geometry.

The stretchability is further demonstrated by reexerting strain on the fully released germanium nanomembrane/PDMS system on a homemade mechanical stage. As the tensile strain is increased gradually, the morphology of the germanium wrinkles changes accordingly, and it reverts to the same configuration as that in the case with the same strain value during the former releasing process. When the stretching strain is increased to 10%, the germanium wrinkles become completely flat similar to the initial state when the germanium nanomembrane is originally transferred to the prestrained PDMS substrate. The wrinkled germanium nanomembrane shows excellent reversibility and can easily restore to any configuration during stretching or release.

The stretchable capability and the reversibility of germanium wrinkles are further confirmed by the variations of L_{edge} [defined as the distance from the free edge to the

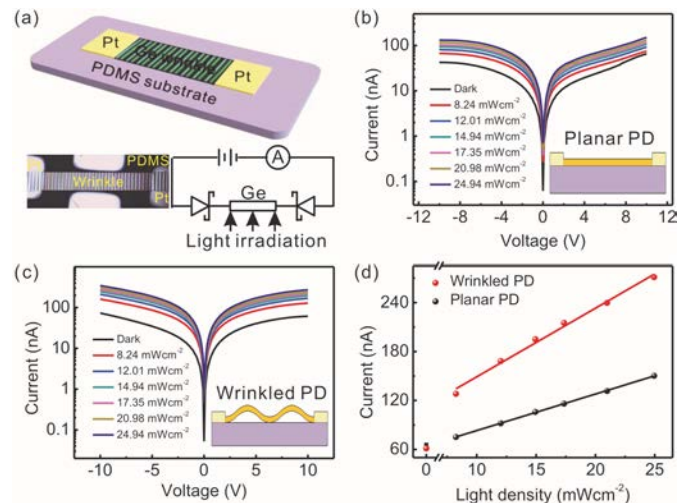


Fig. 3. Optoelectronic properties of photodetective devices fabricated on wrinkled germanium nanomembrane. (a) Schematic (top) and optical microscopic image (bottom left) of structure, an equivalent circuit (bottom right) of the wrinkled PDs, the width and length of the channel are 40 and 250 μm , respectively. (b)–(c) I - V characteristics of PDs fabricated on the planar and wrinkled germanium nanomembranes with or without light irradiation at room temperature. (d) Relationship between the photocurrent and light intensity for the two types of PDs.

midpoint between the first peak and valley nearest to the edge [32], as shown in Fig. 2(b) (inset)], which depends on the strain value during stretching (red circle dots) or releasing (black square dots) process, as shown in Fig. 2(b). For a certain strain of PDMS, the value of L_{edge} obtained from the releasing process is almost the same as that achieved from the stretching process, which verifies the reversibility of germanium wrinkles. Notably, the value of L_{edge} decreases monotonously as the release proceeds (black square dots), which indicates that the initial wrinkles are formed at the middle of the strips. Opposite variation tendency of L_{edge} (red circular dots) can be observed for stretching process as shown in Fig. 2(b), which reveals that the reversion of germanium wrinkles to planar nanomembrane starts from two edges of strips.

C. Flexible Photodetectors Based on Wrinkled Germanium Nanomembranes

The wrinkled germanium nanomembrane can be further constructed into a lateral metal–germanium–metal heterojunction to build wrinkled MSM PDs (wrinkled PD), as shown in Fig. 3(a). For comparison, the counterparts of flexible MSM PDs based on planar germanium nanomembranes (planar PD) without wrinkle geometry are also fabricated (not shown). Platinum is chosen to form the Schottky contact with germanium wrinkles, since the Schottky barrier height between platinum and germanium is thermally stable [35]. A white light source is utilized to characterize the optoelectronic properties of MSM PDs [36], [37], and its power has been precalibrated by a laser power meter [38]. The optoelectronic responses of the planar and wrinkled MSM PDs are assessed by the current–voltage (I - V) characteristics with or without light irradiation, as shown in Fig. 3(b) and (c), respectively. In the absence of light irradiation, a relatively small current (dark current density $\sim 0.4 \text{ mA/cm}^2$ at 5 V) flows through both planar

and wrinkled PD, which indicates good Schottky contact between Pt and germanium. However, under light illumination, the currents increase drastically due to the increase of the photon-generated carrier density [39]. The photocurrents of two types of PDs at the bias of 10 V change linearly with the light intensities, as summarized in Fig. 3(d). From the data exhibited in Fig. 3(d), the responsivities (R) of two types of PDs, expressed as $R = \Delta I / (A * P)$, can be calculated. ΔI is the photocurrent change, which is defined as the difference between the currents with and without illumination, A is the active area, and P is the laser power density. For wrinkled and planar photodetectors biased at 10 V, the responsivities are calculated as 85 and 35 mA/W, respectively, which imply that the germanium-based PD with wrinkled structures exhibits enhanced efficiency of optoelectronic interactions.

Previous studies have demonstrated that the wrinkled or buckled structures can enhance the light extraction efficiency [40], [41], or reduce the reflectance by light trapping effect [42], [43], or scattering effect [44], and thus improving the optoelectronic interactions [45]. In order to investigate the mechanism of the enhanced responsivity for the germanium wrinkled photodetectors, finite-element simulations are performed to explore the light absorption of both types of germanium nanomembranes, i.e., planar nanomembrane and wrinkled nanomembrane. For the finite-element simulation, the thickness of germanium nanomembrane is 50 nm, and the wavelength (λ) and the amplitude (A) of germanium wrinkles are extracted from the experimental results, i.e., $\lambda = 6.12 \mu\text{m}$ and $A = 460 \text{ nm}$, respectively. The planar incident lights with the wavelength ranging from 400 to 800 nm are chosen to illuminate both the planar and wrinkled germanium nanomembranes from the top. Fig. 4(a) shows the cross-sectional view of the electric field distributions for planar germanium nanomembrane (left) and germanium wrinkle (right) on PDMS. Typical light scattering can be observed for the germanium wrinkle due to the wavy surface morphology, and thus inducing significant changes in the propagation directions of light. For more details, light absorptions of both the planar and wrinkled nanomembranes are calculated, as shown in Fig. 4(b). A light absorption of germanium wrinkle is distinctly enhanced compared with that of planar germanium nanomembrane. We attribute this enhanced light absorption of germanium wrinkles to three factors: 1) light trapping behavior of wrinkled structures to reduce the reflections [42], [43]; 2) prolonged effective optical path by scattering effect [44]; and 3) interference-effect of air gaps between the wrinkle and substrate [46], which can be confirmed by SEM results [Fig. 1(g)]. Therefore, the enhanced light absorption of germanium wrinkles can induce more photon-generated carriers, which are responsible for the distinct enhancement in the responsivity of wrinkled PDs.

The optoelectronic response rate of the wrinkled PD is evaluated and shown in Fig. 5(a). It reveals that the response time to a pulsed light irradiation exhibits a long-term repeatability in $I_{\text{light}}/I_{\text{dark}}$ (currents through the source and drain when the light is ON or OFF) at a bias of 10 V. A typical cycle under switched light irradiation is magnified in Fig. 5(b) to clearly show the currents in the light-ON and light-OFF states, and the

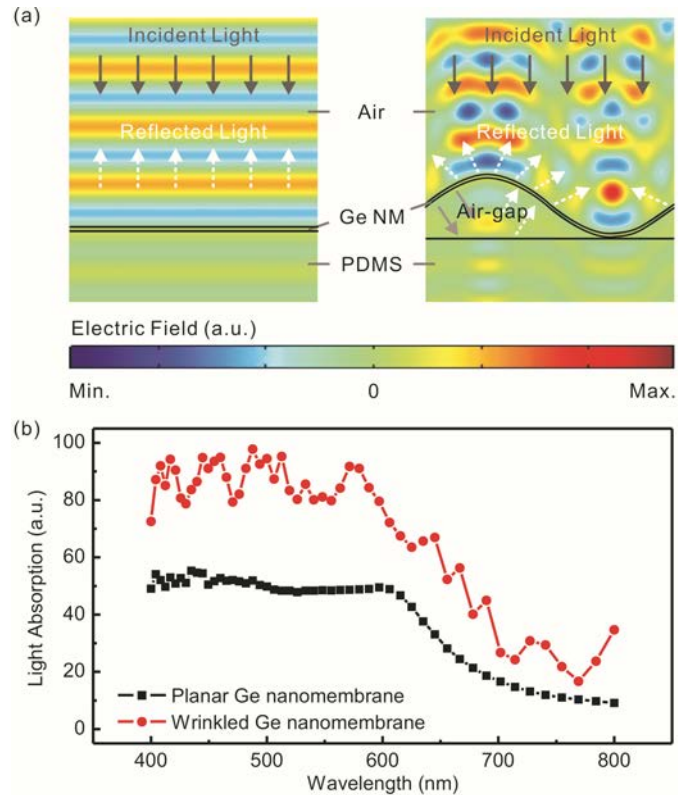


Fig. 4. Simulated light absorptions of the planar and wrinkled germanium nanomembranes. (a) Cross-sectional views of the electric field distributions for the planar and wrinkled germanium nanomembranes. (b) Simulated results of the absorption for the planar and wrinkled germanium nanomembranes in visible light region.

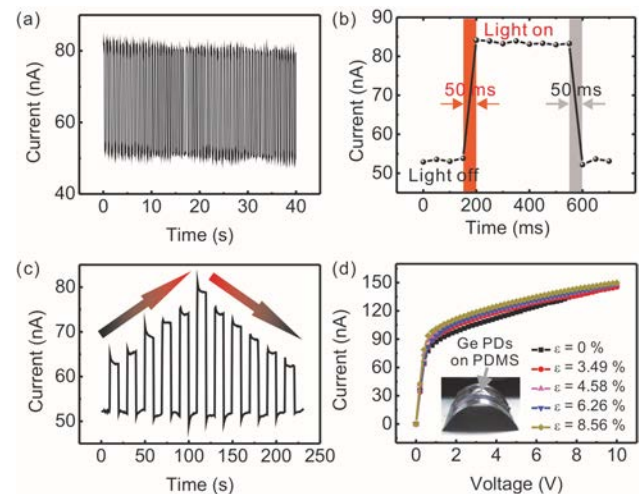


Fig. 5. (a) and (b) Optoelectronic response of the wrinkled PD under pulsed light irradiation at a constant light intensity. (c) Optoresponse of the wrinkled PD under pulsed light irradiation with increasing/decreasing intensity (shown by arrows). (d) I - V characteristics of the PD under various bending-induced strains, and the optical image of the bent germanium PD on PDMS is shown as inserted.

step rise and fall edges in the currents exhibit an optoresponse with a response time of 50 ms. The optoresponse under pulsed light irradiation with variable intensity is examined and summarized in Fig. 5(c). The photocurrent increases

gradually with the light intensity and reaches the maximum when illuminated with the maximum light intensity. As the light intensity is reduced, the photocurrent drops accordingly. Notably, for light irradiation with the certain intensity, the corresponding photocurrent always keeps constant, indicating the good repeatability of wrinkled PD device. Since wrinkles or buckles have been demonstrated to successfully accommodate a vast amount of compressive or tensile strain [13], [29], [30], [44], the wrinkled germanium PDs exhibit superior tolerance to external mechanical bending which generates considerable tensile strain, as shown in Fig. 5(d) (inset). The I - V characteristics of the wrinkled PD without mechanical bending ($\varepsilon = 0\%$) are also monitored after each bending for comparison as shown in Fig. 5(d). Variations in the photocurrent in the wrinkled PD can be barely observed when the external mechanical strain is varied from 0% to 8.6%. Still, the bending capability bodes well for the application of the wrinkled PD device in stretchable/wearable electronics.

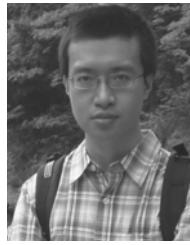
IV. CONCLUSION

In summary, a wrinkled germanium nanomembrane with a wavy topography is fabricated onto a prestrained elastomeric substrate by pattern transfer. The wrinkled Ge nanomembrane can accommodate large external tensile or compressive strain while possessing excellent stretching capability and reversibility. Wrinkled PDs have been demonstrated to exhibit better optoelectronic responsibility than that of planar PDs. Compared with other PD devices fabricated on flexible substrates, the wrinkled PD device shows a high response rate, stretching capability of up to 8.56%. The germanium nanomembrane with a wrinkled configuration is promising for stretchable light sensors or photodetectors in flexible/wearable optoelectronics.

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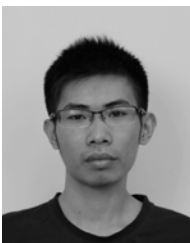
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