Single-Crystalline Silicon-Based Heterojunction Photodiode Arrays on Flexible Plastic Substrates

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***Abstract*—A silicon-based photodiode array was fabricated on a flexible polyethylene terephthalate substrate using a transfer printing technique. A heterojunction structure composed of a 15-nm-thick highly doped hydrogenated amorphous-silicon (n^+ a-Si:H) layer and a 3-μm-thick p-type single-crystal silicon (p c-Si) membrane layer was adopted as the active layer of the flexible photodiode. The highly ordered photodiode array formed on the flexible substrate exhibited superior stability in electrical properties under bent conditions with no mechanical deformation. The variation of the spectral quantum efficiency (QE) under short--wavelength light illumination ($\lambda \leq 580$ nm) was in excellent agreement with that of a heterojunction photodiode composed of a-Si:H and a bulk c-Si substrate. Relatively low QE values were observed under longer wavelength ($\lambda \geq 600$ nm) illumination due to the finite thickness of the active layer. The C–V measurement results of the fabricated photodiode array were in accordance with the abrupt junction model. A closer inspection of the junction area of the device using high-resolution cross-sectional transmission micrograph exhibited an interface depth of 2 ± 0.5 nm, which is unavoidable in plasma-enhanced a-Si:H deposition processes.**

***Index Terms*—Flexible device, heterojunction, silicon-based photodiode, transfer printing.**

I. INTRODUCTION

FLEXIBLE or bendable electronic devices fabricated on plastic substrates have received significant attention as candidates for use in photodetectors, solar cells, displays, and transistors due to their light weight, low manufacturing cost, and folding capability at uneven surfaces in diverse applications [1]–[5]. Polymer or organic materials have been used widely as active layers in these devices since they are compatible with plastic substrates that can endure rather low thermal budgets in their fabrication processes [6], [7]. However, organic-based flexible devices have a certain limitation in the optical performance; in the case of organic semiconductors (e.g., poly(3-hexylthiophene) and poly[2-methoxy-5-(2′-ethylhexoxy)-1,4-phenylenevinylene]), its light absorption and quantum efficiency (QE) in the range of visible light (380–750 nm) are much smaller than those of inorganic semiconductors due to its large band gap (> 2 eV) [8], [9]. To circumvent these restrictions and realize high-performance flexible electronics, printing techniques that allow the relocation of inorganic single-crystalline-silicon (c-Si) thin membranes on flexible substrates have been researched extensively [10]–[12]. In c-Si devices, adequate thickness of the active layer in the range of few tens of micrometers is required to obtain sufficient absorption of ambient light for high photon-conversion efficiency. However, this thickness requirement for the rigid inorganic active layer conflicts with its ability to sustain mechanical stability under bending conditions [13]. Thus, to obtain a flexible device, the absorption ability has to be sacrificed somewhat in exchange with the thickness of the active layer.

In a c-Si based photodiode, heterojunction structure with a hydrogenated amorphous-silicon (a-Si:H) layer has been receiving great attention due to its highly efficient, simple, and cost-effective fabrication process [14]–[16]. The related research has been focused on the carrier transport mechanisms and the electrical characteristics of a-Si:H/c-Si structure in various applications including solar cells, photodetectors, and bipolar transistors [17]–[19]. However, none has yet reported on the fabrication of a flexible photodiode based on this a-Si:H/c-Si heterojunction system.

In this paper, the challenge of making the active layer thinner to the extent of being durable in a bended state was addressed using an a-Si:H/c-Si heterojunction photoarray printed on a flexible polyethylene terephthalate substrate. The 3-μm-thick active layer was detached from the top of a silicon-on-insulator (SOI) wafer by a hydrofluoric acid (HF) wet-etching process and transferred onto a plastic substrate using an elastomeric polydimethylsiloxane (PDMS) stamp. The relocated array of cells was highly ordered with no mechanical deformation. The device characteristics were extracted from the I–V and C–V measurements. High-resolution cross-sectional transmission electron microscopy (HR-XTEM) was utilized to investigate the interface of the a-Si:H/c-Si heterojunction. The photodiode array assembled on the plastic substrate exhibited durable flexibility with excellent electrical performance.

II. EXPERIMENTAL PROCEDURES

Fig. 1(a)–(f) illustrates schematically the procedures used to obtain the a-Si:H/c-Si heterostructure thin-film photodiode array on a plastic substrate. First, the polished SOI wafers with a 3-μm-thick top layer of p-type silicon and a buried oxide of

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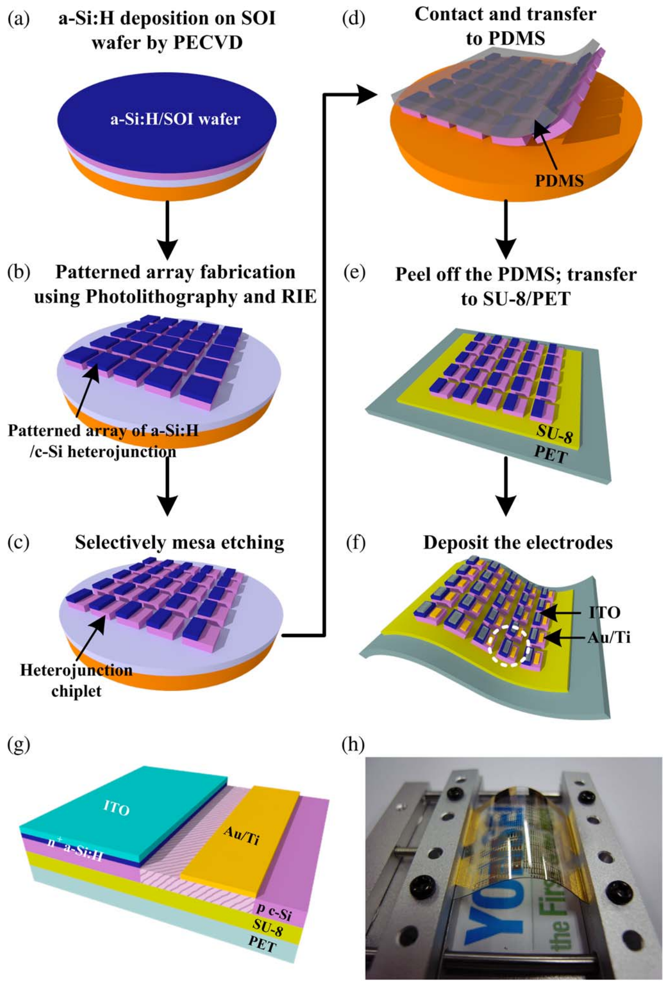

Fig. 1. (a)–(f) Schematic illustration of the process used to fabricate the heterojunction photoarray on a flexible substrate. (g) Perspective schematic of the fabricated a-Si:H/c-Si photodiode chiplet. (h) Photograph of the fabricated photo array on a bent flexible PET substrate.

200 nm were prepared. They were cleaned at 70 °C for 10 min in an RCA solution (1:1:5(v/v/v) $NH_4OH/H_2O_2/H_2O$), followed by dipping in a buffered-oxide-enchant solution for 30 s to remove the native oxide. To obtain heterojunction active layers, the 15-nm-thick highly doped n-type (n^+) a-Si:H thin films were deposited on the SOI wafers using a plasma-enhanced chemical vapor deposition (PECVD) system in a gas mixture of SiH_4 (20 sccm) and PH_3 (1% diluted in 99% H_2 with 10 sccm) at the setpoint temperature of 350 °C [see Fig. 1(a)]. The patterned arrays of a-Si:H/c-Si heterojunction structure were fabricated via photolithography combined with reactive ion etching (RIE) using SF_6 and O_2 mixture [see Fig. 1(b)]. Then, the a-Si:H layer was mesa-etched selectively with the dimensions of 200 μm (width) × 250 μm (length) to expose the p-type (p) c-Si for the individual front-side contact, as shown in Fig. 1(c). To separate the heterojunction active layer from the mother wafer, the samples were immersed in aqueous HF with a concentration of 49%–51% to etch the buried oxide. After the etching process was carried out, the photodiode chiplets in the array form were settled down with maintaining their initial positions on the mother substrate via Van der Waals' force. Prior to transferring the fully detached chiplet array, the PDMS was cured for 24 h at room temperature using a mixture consisting of a Sylgard 184 base and a curing agent at a volume composition ratio of 10:1 [20]. The PDMS stamp was brought into contact with the top surface of the detached arrays, and they were peeled off completely from the mother wafer [see Fig. 1(d)]. Separately, the PET films were cleaned successively with acetone, isopropanol, and deionized water. The surfaces of the PET substrates were treated with O_2 plasma at 300 W for 30 s and were spin-coated with an adhesive SU-8 2002 layer. The SU-8/PET sample was prebaked at 95 °C for 5 min; then, the transferred arrays on the PDMS were attached onto the prepared SU-8/PET substrate at 95 °C for 5 min [see Fig. 1(e)]. Then, the photodiode array/SU-8/PET samples were baked at 115 °C for 10 min to ensure cross-linking of the SU-8 bonding material. To fabricate the front-side transparent electrode (n^+ a-Si:H contact), a 100-nm-thick indium–tin–oxide (ITO) layer with a dimension of 150 μm × 200 μm was deposited selectively with a shadow mask on the n^+ a-Si:H layer using a direct-current magnetron-sputter deposition system with SnO_2 (90%) and In_2O_3 (10%) targets. Finally, the 50-nm-thick Au electrode (200 μm × 100 μm) was evaporated thermally on the exposed p c-Si layer [see Fig. 1(f)]. Fig. 1(g) exhibits the perspective view of a single cell with both the ITO and Au/Ti electrodes on the top side (a lateral structure) denoted with the white circle in Fig. 1(f). The lateral structure was adopted in this experiment due to the difficulties in the formation of the back-side electrodes of the proposed flexible photodiodes. Fig. 1(h) shows an image of the fabricated photodiode array on a bent PET substrate.

The I–V characteristics measurements were performed using a HP4145B semiconductor parameter analyzer. The spectral photoresponsivity was characterized using a light source (Oriel Optical System, 500 W Hg-arc lamp) with a monochromator covering the range of 400 to 900 nm. C–V characteristics were measured under dark conditions at 1 MHz using a KEITHLEY 590 C–V analyzer. The interface of the a-Si:H/c-Si heterojunction active layer was investigated using TEM (FEI Tecnai F20).

III. RESULTS AND DISCUSSION

Fig. 2(a) shows the representative semilogarithmic I–V curve of n^+ a-Si:H/p c-Si heterojunction photoarray on a flexible PET substrate without illumination, revealing that the fabricated diode exhibits a good rectifying behavior. The inset in Fig. 2(a) shows $\ln(j_F/j_0)$ as a function of forward-bias voltage, where j_F is the forward-current density and j_0 is the saturation current density. From the inset graph, the diode ideality factor n of the fabricated device was obtained using the following diode equation [8]:

$$j_F = j_0 \left\{ \exp\left[\frac{q(V - j_F R_S)}{nkT}\right] - 1 \right\} \tag{1}$$

where q is the electron charge, V is the applied voltage, R_s is the series resistance of the diode structure, k is the Boltzmann's constant, and T is the absolute temperature. The calculated n in the low-level injection region was 1.44. The deviation of n from unity may be attributed to the recombination of

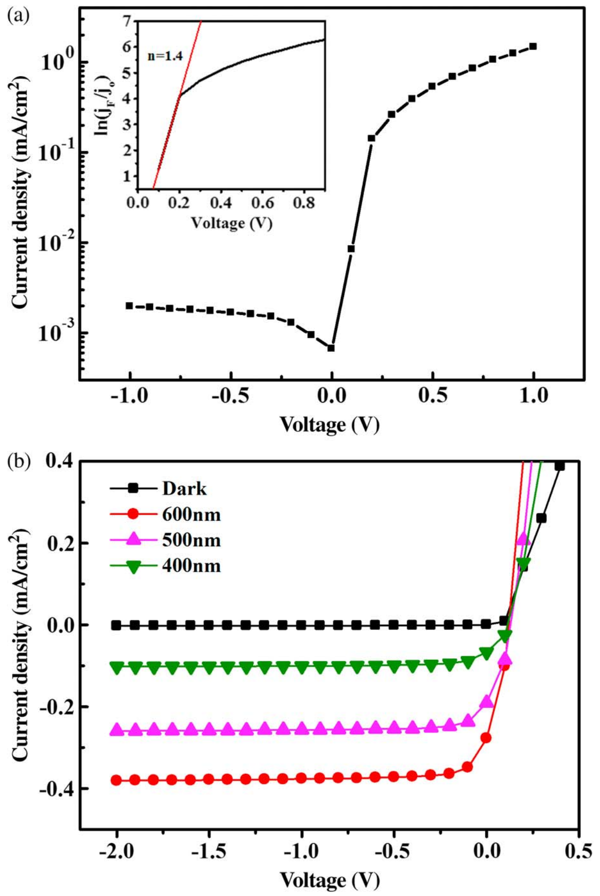

Fig. 2. (a) Semilogarithmic I–V characteristics of a-Si:H/c-Si heterostructure under the dark condition. The inset shows $\ln(j_F/j_0)$ as a function of forward bias at the low-bias regime. (b) I–V characteristics of the a-Si:H/c-Si flexible photodiode array as a function of the illumination wavelength. The incident optical power was normalized at 1 mW.

electrons and holes in the space-charge region. R_s was also obtained in the high-forward-bias region using (1), which was a considerably high value of $\approx$400 kΩ. The R_s of the fabricated device is composed of the contact resistance between the ITO and n$^+$ a-Si:H, the resistance of the n$^+$ a-Si:H layer, the resistance of the depletion region, the resistance of the bulk p c-Si layer, and the contact resistance between p c-Si and Au/Ti electrode. It can be expected that the high value of R_s was originated from an additional resistive part with dimensions of 200 μm $\times$ 170 μm $\times$ 3 μm (width $\times$ length $\times$ height) resulting from the use of a lateral structure, i.e., indicated as a shaded region in Fig. 1(g), which was estimated to be approximately 45 kΩ. In addition, the poor contact between the p c-Si and the Au electrode could also contribute to this large R_s; their interface could be damaged during the RIE process to expose the front-side contact area, which may cause a high contact resistance, and the adequate postannealing treatment after the contact formation in the fabricated device was limited due to the low thermal budget of the PET substrate.

Fig. 2(b) shows the I–V characteristics of the n$^+$ a-Si:H/p c-Si heterojunction photoarray on a flexible PET substrate under illumination with different wavelengths λ. The photodiodes were biased up to -2 V under dark and illumination conditions with wavelengths of 400, 500, and 600 at room temperature. The incoming power of the light illumination with various wavelengths was normalized at 1 mW, and the lights were projected at the front side of the fabricated photodiode arrays. Regardless of the incident wavelength, all of the reverse currents under illumination increased proportionally as a function of $V^{1/2}$ in the low bias region from 0 to -0.5 V and were nearly saturated afterward. The photocurrents under different illumination of λ at the -2 V condition were 0.1 mA/cm^2 at 400 nm, 0.26 mA/cm^2 at 500 nm, and 0.38 mA/cm^2 at 600 nm. Such variation in the photocurrents with varying illumination of λ could be attributed to the fact that the photon flux changes with λ even when the illumination intensity was normalized. As shown in Fig. 2(b), the I–V characteristics under the different light illuminations are not the shifted replicas of the dark I–V curve, which might be originated from the changes in the R_s. Due to the additional resistive part in the proposed lateral structure, the fabricated device performance is more dependent on the variations of R_s compared with that of the vertically structured devices. The values of R_s are closely related to the amount of carrier concentration, which could be affected by the amount of light absorption in the fabricated device [21]. In our case, the light absorption is maximized when illuminated by light with λ of 600 nm (will be discussed in detail later, shown Fig. 3) and accordingly, the value of R_s is minimized, leading to a dramatic increase in the curvature of the I–V curve at the forward-bias region. The current in the dark remained almost constant with increasing reverse bias voltage, indicating that the flexible photodiode is well fabricated without any degradation even throughout the transfer printing process.

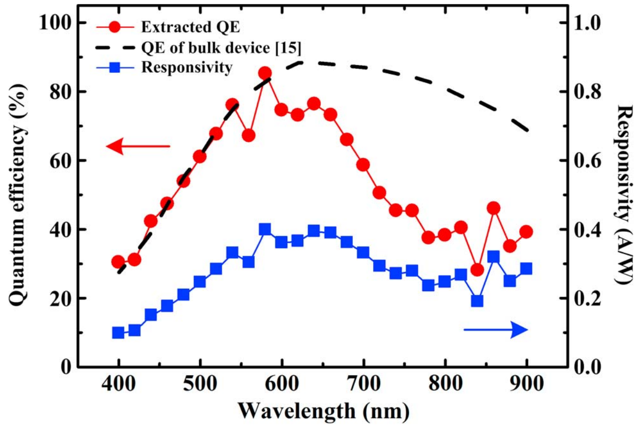

Fig. 3. Extracted spectral QE and corresponding responsivity as functions of the wavelength of the incident photons. For comparative analysis, the dashed line represents the data set in [15], showing the QE of a 10-nm-thick a-Si:H/bulk c-Si device.

Fig. 3 shows the QE and the corresponding photoresponsivity of the transferred photoarrays as the functions of λ of the illumination source, which ranges from 400 to 900 nm. The extracted QE increased nearly linearly from 30% for $\lambda =$ 400 nm to 85% for $\lambda = 580$ nm before decreasing to 39% for $\lambda = 900$ nm. A similar tendency can be found in the spectral photoresponsivity as the QE, as expected. The dependence of QE on the wavelength in the short-wavelength regime ($\lambda \leq$ 580 nm) can be related to the probability of recombination of generated electron–hole (e–h) pairs. Due to the high absorption

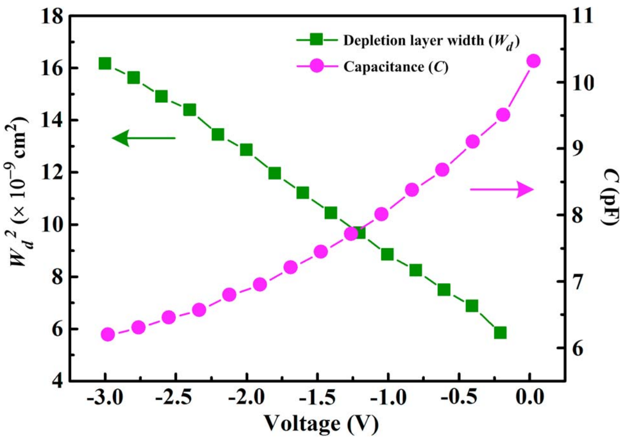

Fig. 4. Square of the depletion layer width W_d^2 and capacitance C as the functions of the applied voltage V at 1 MHz.

coefficient of silicon for short-wavelength incident photons, most of the photons are absorbed near the surface, resulting in the generation of e–h pairs. However, since the top layer of our fabricated structure is an a-Si:H film, the defect density can be expected to be very high; thus, a considerable amount of the generated e–h pairs are subject to recombination, leading to a low QE value. In addition, the use of a polished SOI wafer in the heterojunction structure, resulting in the reflection loss, could contribute partially to this low QE. The highest QE of the transferred photoarrays was achieved at $\lambda = 580$ nm. This can be ascribed to the fact that the thickness of the active layer of the fabricated photoarray was 3 μm. For a comparative analysis, the appropriate data set from the work of Borchert *et al.* [15], who investigated the spectral QE of a heterojunction photonic device in a c-Si bulk wafer while varying the thickness of the a-Si:H layer, was retrieved. The dashed line in Fig. 3 indicates the QE obtained from their 10-nm-thick a-Si:H/bulk c-Si heterojunction in [15], which is well consistent with our experimental results in the short-wavelength region. The estimated penetration depth of the monochromatic light with $\lambda = 600$ nm in the c-Si is approximately 3 μm [21], which is consistent with the thickness of the active layer of our photoarray. Thus, at wavelengths below 600 nm, the electrical characteristics of our fabricated thin-film photoarray are similar to those of a bulk silicon device. However, with illumination with longer wavelengths ($\lambda \geq 600$ nm), the QE of the photoarray was lower than that of the bulk silicon device. Such differences in the QE can be attributed to the fact that the thickness of our active layer was smaller than the penetration depth of longer wavelength light ($\lambda \geq 600$ nm) in silicon. Thus, a substantial amount of the long-wavelength incident light penetrated through the silicon film of the photodiode array, causing less e–h generation, which in turn led to a considerable decrease in the QE and photoresponsivity.

Fig. 4 exhibits the variations of the square of the depletion layer width W_d^2 and the capacitance of the fabricated flexible photodiode array when varying the applied voltage from -3 to 0 V. The C–V measurements were carried out at 1 MHz. This frequency is high enough to neglect the dielectric relaxation process in a-Si:H [22]. Thus, the total capacitance of the device can be regarded as the series capacitance of the a-Si:H layer and the c-Si layer. The capacitance of the a-Si:H layer may be a constant value, and the capacitance of c-Si depends on the depletion width of the junction, which varies with the applied voltage. Assuming that this junction follows the abrupt heterojunction model, we can express the dependence of W_d on the applied voltage as follows [23]:

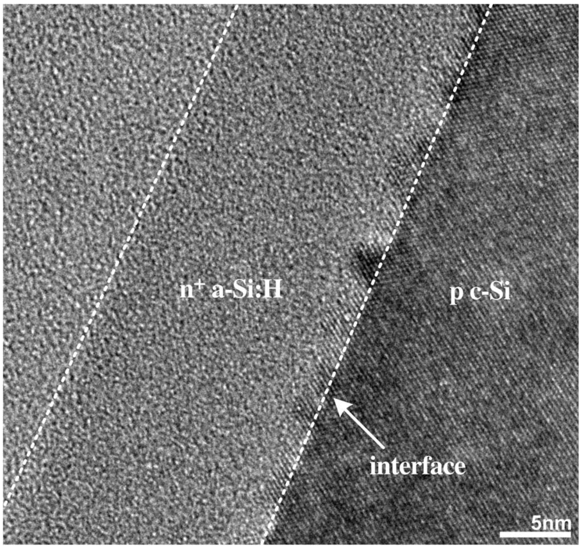

Fig. 5. HR-XTEM micrograph of the a-Si:H/c-Si interface.

$$W_d^2 = \left(\frac{\varepsilon_0 \cdot \varepsilon_s}{C} - L\right)^2 = \frac{2\varepsilon_0 \varepsilon_s N_I}{q N_A (N_I + N_A)} (V_D - V) \qquad (2)$$

where L is the thickness of the a-Si:H layer, ε_0 is the free-space permittivity, ε_s is the dielectric constant of a-Si:H and c-Si, q is the electronic charge, C is the capacitance of the depletion layer, N_I is the effective density of the donor-like states in a-Si:H, N_A is the doping concentration of c-Si, and V_D is the diffusion voltage. In particular, in the case of our fabricated n$^+$ a-Si:H/p c-Si heterojunction, the carrier density and effective donor-like states of the n$^+$-layer are much higher than that of the p-layer. Hence, (2) can be simplified as follows [24]:

$$W_d^2 = \frac{2\varepsilon_0 \varepsilon_s}{q N_A} (V_D - V). \qquad (3)$$

As shown in (3), the magnitude of N_A can be deduced graphically from the slope of the W_d^2 versus V curve. The estimated N_A value obtained from the slope of the plot in Fig. 4 is 3.43×10^{15} cm^{-3}, which is almost identical to the doping concentration of p c-Si in this paper ($2.3 \pm 1.1 \times 10^{15}$ cm^{-3}). As shown in Fig. 4, W_d^2 has a linear relationship with the applied voltage, indicating that the abrupt heterojunction model is applicable to our fabricated system [22]. Consequently, it was confirmed that the fabricated junction was electrically abrupt.

Fig. 5 represents a typical HR-XTEM image of the a-Si:H/c-Si heterojunction interface of the fabricated photoarray showing a slightly rugged interface. Such an uneven interface can be caused generally by either the amorphization of the c-Si by plasma damage or the epitaxial growth of the c-Si into the a-Si:H matrix during the PECVD process of the thin a-Si:H layer [25]–[27]. In our case, the faintly rugged interface may have been formed due to the epitaxial growth of c-Si, which was facilitated by the PECVD process with sufficiently high deposition temperature for the epitaxial growth of c-Si. This irregular interface played a substantial role in the degradation of the electrical properties of the fabricated devices in earlier reports [28], [29] as the interface contained much structural defects. In contrast, it seems that the amount of local defects originated from the interface in our photoarrays was comparatively small, and thus their effect on the electrical properties could have been minor. As shown in Fig. 5, the thickness of the epitaxial grown c-Si layer in the interface area was approximately 2 ± 0.5 nm, which was relatively small compared with those of the previously reported structures [25], [27], inferring that the electrical degradation of our fabricated photodiode by interfacial defect states might also be comparably small. Thus, combining these results, we conclude that the heterojunction photoarray was well fabricated with minimal degradation of the electrical properties by the deformed heterojunction interface.

IV. CONCLUSION

The device performances of a transfer-printed a-Si:H/c-Si heterojunction photoarray on a PET substrate were investigated using I–V and C–V measurements and responsivity tests under illumination at different wavelengths. The transferred photoarray chiplets were highly ordered and did not show any mechanical deformation after repeated folding–unfolding operation. The fabricated photodiode exhibited a low level of dark current and high photosensitivity. The decreased photoresponsivity with illumination with a long wavelength can be related to the light penetration depth, which exceeds the thickness of the active layer. By combining the HR-XTEM analysis and C–V measurement results, we demonstrated that the fabricated heterojunction active layer was electrically abrupt with an interface with small roughness, which was applicable in various flexible electronic applications.

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