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360° omnidirectional, printable and transparent photodetectors for flexible optoelectronics

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Flexible optoelectronics that can be bent, wrapped, and stretched have attracted interest for wearable and mobile applications. In this work, we demonstrate a transparent 360° omnidirectional photodetector (PD) that can be stretched and wrapped around flexible or curved substrates. By embedding interlaced ZnO and Ag nanowires (NWs) in thermoplastic polyurethane via inkjet printing, the device featured > 75% transmittance in the visible region, showing high photoresponsivity and response time (10–30 A/W and 0.8 s, respectively). Moreover, the flexible PD performs well under deformation (only 9% decay in the photocurrent under 60% strain and 8% loss when the device is bent at a radius of 5 mm), which allows it to be readily applied on curved surfaces, such as skin or optical fibers. This study opens the door for the development of flexible optoelectronics that could be implemented in fiber optics, wearable electronics, self-powered systems, bio-signal monitors, and epidermal electronics.

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INTRODUCTION

Optoelectronic devices have played an important role in information transmission, energy harvesting, environmental monitoring, and biomedical applications.^{1–7} To integrate these applications even closer to our daily lives, it is necessary to develop wearable and mobile systems for the next-generation of electronics, such as flexible displays, wearable photonic textiles, self-powered systems, and biological signal monitoring.^{5,6} In order to achieve these applications, the entire electronic system, including the active layer, electrodes, and the circuits,⁸ must be mechanically flexible and stretchable in order to accommodate comfortable body motion.

One strategy to achieve these goals is to embed rigid devices into flexible substrates as hosting matrices. For example, researchers have demonstrated that conventional inorganic devices made on plastic substrates can be flexible and conformable on curvilinear surfaces, such as human skin.^{9,10} Nonetheless, stretching such devices remains a challenge because the large difference in Young's modulus between the rigid and flexible materials can cause detachment at the interface. Moreover, under high deformation, the rigid devices cannot maintain their functionality because the continuous films of active or electrode layers break apart, thus degrading device performance and limiting practical applications of wearable electronics. Although delamination and broken films can be avoided by minimizing the device size or optimizing the device-to-device distance, it also limits the active area, which decreases optical absorption and lower space utilization.^{10,11}

To overcome these challenges, nanomaterials are a promising option that not only provides desired functionalities but also offers structural advantages that surmount many limitations faced by rigid devices.^{12–21} Composites of nanomaterials and polymer

networks, in particular, have distinct advantages as they retain the electronic properties of the nanomaterials as well as provide stability under deformation, including stretching and bending. Moreover, the high transparency of nanomaterials is another desired feature for future optoelectronic applications that is not readily achieved using conventional bulk semiconductors. Noteworthy examples include stretchable electrodes made using metal nanosheets, graphene, and carbon nanotubes.^{16,22–24} Researchers have also demonstrated strain sensors using Ag nanowires (NWs) and graphene.¹⁷ Most studies use nanomaterials as the electrodes or other single electronic components. However, very few have reported a fully integrated all-flexible optoelectronic device.

The ability to detect light from an oblique angle is another need for optoelectronic applications, such as photovoltaics, omnidirectional cameras, optical tracing systems, and optical field measurements.²⁵ To improve the omnidirectional detection capabilities of optoelectronic devices, researchers usually employ antireflection layers, such as NW arrays and resonant plasmonic and metamaterial structures.^{26–28} However, an inherent limitation is that non-transparent planar devices can only detect incident light from one-half of ambient space (*i.e.*, the front 180° field of view). To detect light behind the device (in the rear 180° field of view), it is necessary to use sophisticated lenses, prisms, and other optical components, which makes the system cumbersome.²⁹

Here we report a fully transparent, stretchable, and wrappable photodetector (PD) using an interlacing NW network embedded in a flexible polymer substrate by means of printing approaches. The device was constructed using ZnO NWs as the photo-active channels interlaced with Ag NW electrodes in a well-ordered cross-junction array. The entire device is completely flexible and transparent with over 75% transmittance in the visible region, thus enabling 360° omnidirectional photodetection. Incident light from

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ambient angles showed only 78% variance in responsivity change, which is in contrast with traditional PDs that can only detect light from the front half of ambient space. Due to the high number of intersections of the interlacing NWs, the PD is able to function under significant mechanical deformation, including bending around a radius of <5 mm and stretching at over 60% strain. This combination of omnidirectional detection and flexibility allows us to wrap the device on curved surfaces while still maintaining its high functionality. With wrappable features, the PD can better detect point light sources without suffering the inverse-square decay compared to planar or non-wrappable photodetection devices. We can readily demonstrate this advantage by measuring the leakage of light from a damaged optic fiber. The resulting PD displays transparency, high deformability, and omnidirectional detectability, which presents key advantages for the future-generation optoelectronics.

RESULTS AND DISCUSSION

Figure 1a shows the proposed stretchable and transparent NW PD. We constructed the device using an interlacing array of ZnO NWs as the active material and Ag NW as the electrodes, both embedded in a thermoplastic polyurethane (TPU) substrate. The electrodes and active materials were deposited via inkjet printing, which allows the pattern of the stretchable PD to be digitally controlled. Instead of using other conventional polymer materials, we employed TPU as a supporting substrate because it features an ideal combination of ultraviolet (UV)-transparency (transmittance > 80% at 365 nm), high elasticity (maximum strain ~ 900%), and high abrasion resistance.³⁰

The procedure for fabricating the NW networks that compose the flexible PD is shown in Fig. 1b. To pattern the device, we first printed Ag NWs in an array of square features on a glass substrate using a commercial inkjet printer. Then we printed a continuous layer of ZnO NWs over the patterned Ag NW electrodes with the same printing system, followed by an annealing procedure to fully remove the ink solvent (the minimum feature size achievable with our inkjet printing system was 50 μm). Scanning electron

microscopy (SEM) images show the two NW networks intersecting with each other in a patterned array (Fig. 1c, d). Next, liquid-phase TPU elastomer was degassed and uniformly spin-coated over the cross-patterned Ag and ZnO NWs. After curing, we peeled the TPU layer off the glass substrate, transferring with it the embedded NW pattern.

This fabrication strategy provides three advantages. First, the device is transparent since it is constructed entirely of nanomaterials and polymer, as shown in Fig. 1e. Second, the network of metal and semiconducting NWs provides a high number of intersections of the two NW networks, which improves the contact resistance and stretchability of the device.^{31–33} We observed that the resulting PD exhibits excellent stretchability and durability against mechanical deformation. Finally, the entire fabrication process of the device is simple, scalable, and low-cost, making large-scale production and practical applications feasible. Meanwhile, the device is compatible with other printable electronics, such as inkjet-printed memory and printable sensors.^{31–33}

We first characterized the basic optoelectronic properties of the printed NW network PD. The *I/V* characteristics and photore sponses of the device are shown in Fig. 2a (a logarithmic plot is shown in Supplementary Information). The dark current (off state) of the device is lower than 100 pA at 5 V due to the multiple junctions within the entangled ZnO NW network. Under UV illumination, the photocurrent (on state) increases by 2 orders of magnitude, corresponding to a responsivity of 6.4 A/W (see Methods). Note that the channel area is used as the UV exposure area for calculating the incident power as well as the responsivity. To characterize the detection wavelength region, we measured the device's spectral response (Fig. 2b). The photoresponse starts at ~370 nm, which corresponds to the wide bandgap of ZnO (3.37 eV).^{34,35} Note that the device shows no response to visible light (Fig. 2a, b), thus exhibiting visible-blind characteristics that are beneficial for making visible-transparent devices. Figure 2c presents the dependence of the device's responsivity on the incident UV power. As with other previously reported ZnO NW network-based devices, the responsivity of our PD increases as the incident power decreases.^{36–38}

Fig. 1 **a** Schematic of an array of NW PD structures implanted into a flexible TPU substrate. **b** Fabrication process of the NW PD device. **c** SEM image of the device geometry. The yellow region (false color) represents the Ag electrodes and the blue region represents the ZnO active material used for photodetection. **d** An enlarged view of (c) showing that both the electrodes and active materials are composed of a network of NWs. **e** A photograph of the transparent PD

Fig. 2 **a** The *I/V* characteristic curves of the NW PD under dark, UV, and visible illumination. **b** The responsivity of the PD at different wavelengths. **c** The dependence of the device responsivity on the incident power. **d** The left panel shows the time-domain photoresponse alternating between on and off states for five cycles at 5 V. The right panel presents the analysis of the response and recovery times

Another feature of the NW PD is its fast response speed. The left panel of Fig. 2d presents the time-domain photoresponse of the NW PD, which was measured by periodically switching on a UV light source (365 nm) with an intensity of 1 mW/cm². The response and recovery times are defined as the time needed for the signal to rise to 80% of the dark current and recover to 20% of the maximum photocurrent, respectively.³⁹ From the analysis in the right panel of Fig. 2d, the device response and recovery times were found to be 0.8 s and 1.6 s, respectively, which is 1–2 orders faster than that of planar ZnO- and aligned NW-based PDs (Supplementary Table S1). The fast photoresponse is attributed to the multiple junctions between the NWs in the interlacing NW network, which has been previously shown to increase the photoresponse recovery speed.⁴⁰ The combined advantages of high photosensitivity with fast response are of practical importance for applications such as optical amplifiers, real-time imaging, and optical communications.

A main feature of the device is its high transparency within the visible light region. The spectral measurements in Fig. 3a, b quantitatively reveal how the TPU, Ag, and ZnO components of the NW PD affect the transparency and absorption of the device. The results show that TPU is an ideal transparent substrate, featuring transmittance of over 85% and absorption of less than 5% within the visible and near UV region (300–900 nm). After the Ag and ZnO NWs were embedded within the TPU, the average transmittance remained as high as 75% within the visible range (400–900 nm), which is comparable to ITO glass (around 80%). The drop in transmittance at wavelengths lower than 400 nm is due to the UV absorption of ZnO NWs.

Note that due to the high UV transparency of the TPU substrate (absorption < 5% from 300 to 400 nm), incident UV light from all directions can reach the ZnO NWs without being blocked, thus realizing the 360° omnidirectional detection capabilities of this device. To demonstrate the angular detection ability, we measured the photoresponse ($\Delta I = I_{\text{photo}} - I_{\text{dark}}$) of the NW PD

by illuminating it with UV light from every angle ($\theta = -180^\circ$ to 180° and $\varphi = 0-360^\circ$, Fig. 3c, d). Here the common spherical coordinate system is used, in which θ denotes the polar angle and φ denotes the azimuthal angle. In Fig. 3f, the photoresponse shows no significant changes as the azimuthal angle was varied since the PD is flat with no structural asymmetry. However, we observed a periodic variation of the photoresponse as a function of polar angle (Fig. 3g). The peak photoresponse occurs when the light is normally incident ($\theta = 0^\circ$) to the device, and the photoresponse decreases with the angle by a factor of $\cos(\theta)$, which is in agreement with the variation of light intensity as a function of polar angle for planar devices. Notably, when illuminated from the rear side of the PD ($-180^\circ < \theta < -90^\circ$ and $90^\circ < \theta < 180^\circ$), the device was still able to detect the UV light because of the high transparency of the TPU substrate and NWs.

To further demonstrate the omnidirectionality of the PD, in Fig. 3e we measured the angular responsivity, $R_{\text{angular}}(\theta, \varphi)$, by considering the incident power $P_{\text{cos}}(\theta)$ at different incident angles:

$$R_{\text{angular}}(\theta, \varphi) = \Delta I(\theta, \varphi) / P_{\text{cos}}(\theta), \quad (1)$$

in which $\Delta I(\theta, \varphi) = I_{\text{photo}} - I_{\text{dark}}$ and P is the normal incident power of the UV light.⁴¹ Using Eq. (1), we determined that the average $R_{\text{angular}}(\theta, \varphi)$ for light coming from the front ($-90^\circ < \theta < 90^\circ$) and back ($-180^\circ < \theta < -90^\circ$ and $90^\circ < \theta < 180^\circ$) of the device was 5.5 A/W and 4.6 A/W, respectively. Further analysis shows that the average $R_{\text{angular}}(\theta, \varphi)$ for incident photons from ambient space was ~78% of the normal-incident responsivity, thus demonstrating the outstanding omnidirectional detection capability of the device.

Since the ZnO and Ag NWs were firmly embedded in the TPU substrate, the adhesion of the NWs to the polymer was greatly improved, which allowed the device to be flexed and stretched to a high degree. Figure 4a, b illustrates the stretching and bending tests of the NW PD, executed at different axes by stretching/

Fig. 3 **a** The transmittance and **(b)** absorption spectra of the different components of the PD device, including the TPU, Ag and ZnO NWs. **c** An illustration of the spherical coordinate system used in this study. **d** The photocurrent measured at different angles of incidence (AOI). **e** $R_{\text{angular}}(\theta, \phi)$ extracted from **(d)**. **f** The photocurrent along θ , in which each point is the average photocurrent from $\phi = 0^\circ$ to $\phi = 360^\circ$. **g** Photocurrent along ϕ , in which each point is the average photocurrent from $\theta = -180^\circ$ to $\theta = 180^\circ$

bending the TPU substrate along the lateral (x axis) and longitudinal (y axis) directions of the active ZnO NW channels. A 365 nm wavelength UV lamp illuminated the device from the top position ($\theta = 0^\circ$). Our results show the PD is capable of stretching and bending while retaining the photocurrent (Fig. 4c, d). As we applied 60% strain to the PD along the perpendicular direction relative to the active ZnO NW channels, the photocurrent decreased by only 9%, which we attributed to the firm interlacing of the Ag and ZnO NW networks in the TPU substrate. These NW networks help to retain the efficient transport of the photo-generated carriers even under high deformation of the device due to the high number of intersecting Ag and ZnO components. However, when we stretched the device by 60% along the axis of the ZnO channels, the photocurrent decreased by 46%, possibly due to the slight detachment of the NWs. In the bending test, the

photoresponse exhibited negligible changes, in which the photocurrent decreased by 8 and 7% while bending the PD device along the x and y, respectively axes (bending radius = 5 mm).

To examine the stability of the device, we measured the retention of photodetectivity after repeatedly deforming the device. After stretching for 100 cycles at 60% strain, the photocurrent retained 95 and 84% of its original value along the x and y axes, respectively (Fig. 4e). Note that we observed no permanent degradation of the device's photodetective characteristics after relaxation of the strain, indicating that the slight detachment of the NW network is recoverable. In addition, the photocurrent maintained 98 and 94% of its original value after bending the device for 100 cycles (bending radius = 5 mm) along the x and y axes, respectively (Fig. 4f). The excellent repeatability

Fig. 4 Stretching and bending tests of the NW PD device. An illustration of the (a) stretching test and (b) bending test executed in two different directions. **c** The normalized photocurrent as a function of strain. The inset shows a series of optical images under different stretching conditions. **d** The normalized photocurrent as a function of the bending radius of curvature. **e** The performance of the PD as a function of stretching cycle with a strain of 60%. **f** The endurance of the PD performance as a function of bending cycle at a bending radius of ~5 mm. Note that the error bars indicate standard deviation of the current measured from >10 different devices

of this flexible PD device could be due to the structural robustness as well as the strong adhesion between the NW networks and the TPU substrate.

Due to the deformability, the NW PD can be readily stretched and twisted without performance degradation (Fig. 5a). In addition, the device can be attached to the body, wrapped on a tube, and stuck to the surface of curved objects with conformal contact (Fig. 5b–d), making it a potential candidate for a component of future-generation flexible optoelectronics.

We also explored the use of the PD for specific applications that require flexibility. For example, the optical time-domain reflectometer (OTDR) is a device used to characterize optical fibers by injecting a series of optical pulses into the fiber and recording the back-scattered light from the same end.⁴² Leaky points in the fiber can be determined by measuring the strength of the return pulses, first integrated as a function of time and then converted to the location along the fiber. However, dead zones, caused by multiple leakage points in close succession or slow attenuations, limit the spatial resolution of the OTDR (Fig. 5f and the Supplementary Information). Since the intensity of leaking light attenuates with distance, it is necessary to place the detector as close to the leakage point in the optical fiber as possible. A new device that could be wrapped around the dead zones in the fiber would help address this problem.⁴³ Combining flexible, stretchable, and transparent features, the NW PD can serve as a complementary instrument for the detection of leaking points of light by physically wrapping it around the optical fiber.

To demonstrate this application, we designed an experiment to detect light on a curved object as shown in Fig. 5e. A UV laser was

coupled onto one end of an optical fiber and the power was measured at the other end. The fiber itself was intentionally broken in the middle (Fig. 5f). A schematic of deadzone of OTDR detection is shown in Fig. 5g. We constructed an array of the PDs along a narrow sheet of TPU, in which the distance between each PD was 1 mm, and wrapped this sheet around the fiber. When the laser was off, the current levels of all the PDs were lower than 1 nA (at 5 V), which demonstrates the off state of the device. However, when the laser was turned on, the PD at $x = 6$ mm showed a peak in the photoresponse, indicating a light leakage point in the fiber where it also happened to have been broken (Fig. 5h). These results show that flexible PD arrays can locate the position of light leaks on a curved surface, which could be advantageous for next-generation optoelectronic applications.

CONCLUSION

In conclusion, we have successfully demonstrated a transparent, omnidirectional, stretchable, and wrappable PD fabricated by inkjet printing. This device has a transmittance of over 75% in the visible region, which engenders the PD with 360° omnidirectional detection capability. The PD can perform under extreme and repeated deformations, including bending (bending radius = 5 mm) and stretching (strain = 60%), due to the unique interlaced structure of the Ag and ZnO NW networks embedded in the polymer substrate. Combining omnidirectional and flexible features, the device can be wrapped on any round surface and maintain its high photodetection performance. This wrapping capability enables detection over curved surfaces, such as the

Fig. 5 **a** The PD being stretched and twisted. **b** The flexible PD device attached to skin, **(c)** wrapped on a light-emitting diode tube (the scale bar is 1 cm), and **(d)** stuck to the surface of a sphere. **e** The experimental setup to detect a leaky point in an optical fiber. **f** An illustration of the PD array wrapped on a fiber and the optical images of the fiber and PD arrays (the scale bar is 1 cm). **g** A scheme describing the dead zones of the OTDR technique. **h** The photoresponse of the PD array under dark and different laser power illuminated conditions

determination of light leakage along an optical fiber. This concept for omnidirectional and stretchable PDs will expand the applications for future wearable electronics, self-powered systems, functional clothes, and epidermal electronics.

METHOD

Device fabrication

Inkjet printing was carried out using a MicroFab JetLab4 system (MicroFab Technologies Inc.). TPU is purchased from Huntsman. The thickness of TPU is in 1–2 mm in thickness. The chemically modified AgNW thin film was transferred to TPU substrates by a casting method. The preparation of NW ink as well as the detail fabrication procedures are provided in the Supplementary Information. We performed morphological studies of the NW PD with a JEOL JSM-6500 field-emission SEM.

Characterization of optoelectronic properties

We characterized the optoelectronic properties of the NW PD using a Keithley 4200 semiconductor parameter analyzer. A 365 nm wavelength UV lamp was used as the UV source. The optical power intensity was varied with a combination of neutral density filters. The responsivity was calculated by $R = (I_{\text{photo}} - I_{\text{dark}}) / P$. The spectral measurements were performed by coupling a halogen lamp to a monochromator (Enlitech QE-R). A He-Cd laser (325 nm) was used to demonstrate the detection of light leakage in an optical fiber. Note that the channel area is used as the UV exposure area for calculating the incident power as well as the

responsivity; meanwhile, no obvious degradation/damage/yellowing of the TPU substrate is observed under UV exposure for over 1 day. The dark current is in the range from 100 pA to 1 nA at 5 V. Photocurrent is in the range from 1 to 10 μ A at 5 V under UV illumination. The variation of the response speed is in the range from 0.6 to 0.8 s and decay time is in the range from 1.5 to 2 s. All the measurements were performed at room temperature under ambient conditions.

Data Availability

All data generated or analyzed during this study are included in this published article (and its Supplementary Information Files) or are available from the corresponding author upon request. The computer code generated during the current study is available from the corresponding author on request.

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AUTHOR CONTRIBUTIONS

Experiments were designed by J-H.H. and performed by D-H.L., H-P.W., S-B.C., Y-C.C., and C-L.W.. G-R.L. and Y-C.L. contributed to the writing of the manuscript.

ADDITIONAL INFORMATION

Supplementary information accompanies the paper on the *npj Flexible Electronics* website (<https://doi.org/10.1038/s41528-018-0032-2>).

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