# Ultrathin In<sub>2</sub>O<sub>3</sub> Nanosheets toward High Responsivity and Rejection Ratio Visible-Blind UV Photodetection

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Photoelectrochemical-type visible-blind ultraviolet photodetectors (PEC VBUV PDs) have gained ever-growing attention due to their simple fabrication processes, uncomplicated packaging technology, and high sensitivity. However, it is still challenging to achieve high-performance PEC VBUV PDs based on a single material with good spectral selectivity. Here, it is demonstrated that individual ultrathin indium oxide (In2O3) nanosheets (NSs) are suitable for designing high-performance PEC VBUV PDs with high responsivity and UV/ visible rejection ratio for the first time. In<sub>2</sub>O<sub>3</sub> NSs PEC PDs show excellent UV photodetection capability with an ultrahigh photoresponsivity of 172.36 mA W<sup>-1</sup> and a high specific detectivity of  $4.43 \times 10^{11}$  Jones under 254 nm irradiation, which originates from the smaller charge transfer resistance ( $R_{ct}$ ) at the In<sub>2</sub>O<sub>3</sub> NSs/electrolyte interface. The light absorption of In2O3 NSs takes a blueshift due to the quantum confinement effect, granting good spectral selectivity for visible-blind detection. The UV/visible rejection ratio of In2O3 NSs PEC PDs is 1567, which is 30 times higher than that of In2O3 nanoparticles (NPs) and exceeds all recently reported PEC VBUV PDs. Moreover, In<sub>2</sub>O<sub>3</sub> NSs PEC PDs show good stability and good underwater imaging capability. The results verify that ultrathin In<sub>2</sub>O<sub>3</sub> NSs have potential in underwater optoelectronic devices.

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# 1. Introduction

Visible-blind ultraviolet photodetectors (VBUV PDs), converting UV signals into electrical signals, are the fundamental building blocks for sensing, space exploration, and communications.<sup>[1-7]</sup> Although conventional solid-state VBUV PDs have been widely investigated based on various wide-bandgap nanomaterials,[8-10] photoelectrochemical-type (PEC) VBUV PDs have attracted ever-growing attention due to their simple fabrication process, low-cost, high sensitivity, and potential applications in underwater optical communication.<sup>[11,12]</sup> Various wide-bandgap materials with different nanostructures have been designed for PEC VBUV PDs,<sup>[13-16]</sup> but single material-based PEC PDs usually show relatively poor photoresponse.<sup>[15,16]</sup> Some strategies have been developed for optimizing the performance of PEC VBUV PDs, such as coating heavy metal nanoparticles<sup>[17,18]</sup> and building het-

erojunctions,<sup>[19,20]</sup> and they inevitably increase complexity and cost. Therefore, it is important to explore more wide-bandgap semiconductors with good optical and electrical properties for building high-performance PEC VBUV PDs.

In<sub>2</sub>O<sub>3</sub> is an important n-type semiconductor with great potential in UV optoelectronic devices due to its suitable bandgap,<sup>[21,22]</sup> good optical and electrical properties,<sup>[23]</sup> and good stability.<sup>[24]</sup> In<sub>2</sub>O<sub>3</sub> nanostructure-based conventional solid-state UV PDs show good UV photoresponse, demonstrating their great potential application in UV PDs.<sup>[21,25]</sup> Recently, our group demonstrated the potential of In2O3 microrods in PEC UV PDs with high responsivity and good stability.<sup>[26]</sup> However, the spectral selectivity of In2O3-based UV PDs should be further improved due to most reported In<sub>2</sub>O<sub>3</sub>-based UV PDs having a bandgap in the range of 2.6-2.8 eV.<sup>[21,25,26]</sup> Labram et al. demonstrated that ultrathin In<sub>2</sub>O<sub>3</sub> films showed a wider bandgap as the thickness decreased to less than 5 nm due to the quantum effect.<sup>[27]</sup> Therefore, it is of great significance to investigate In<sub>2</sub>O<sub>3</sub> with a wider bandgap for designing PEC VBUV PDs with good spectral sensitivity.

In this work, we investigated the photoresponse of ultrathin  $\rm In_2O_3~NSs$  and  $\rm In_2O_3~NPs$ -based PEC VBUV PDs.  $\rm In_2O_3~NSs$  were synthesized by a hydrothermal method with annealing^{[28]} and  $\rm In_2O_3~NPs$  were commercial samples.  $\rm In_2O_3~NSs$  show

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**Figure 1.** Characterization of ultrathin In<sub>2</sub>O<sub>3</sub> NSs. a) XRD patterns. b) TEM images. Inset: corresponding HRTEM image. c) Height profiles. Inset: corresponding AFM image. d) UV–vis absorption spectra. Inset: corresponding Tauc curve of ultrathin In<sub>2</sub>O<sub>3</sub> NSs as direct bandgap semiconductors.

ultrathin features with thicknesses in the range of 3.5-4.5 nm, while the size of commercial  $In_2O_3$  NPs is  $\approx 40-60$  nm. Compared with In<sub>2</sub>O<sub>3</sub> NPs, the light absorption of ultrathin In<sub>2</sub>O<sub>3</sub> NSs shifts to the visible-blind region due to the quantum confinement effect.<sup>[27]</sup> In<sub>2</sub>O<sub>3</sub> NSs-based PEC UV PDs show ultrahigh responsivity of 172.36 mA W-1 and excellent specific detectivity of  $4.43 \times 10^{11}$  Jones under 254 nm illumination at a bias voltage of 0.4 V, respectively, surpassing all reported ultrathin nanomaterial-based PEC UV PDs. The excellent UV photoresponse is attributed to the smaller  $R_{ct}$  between  $In_2O_3$ NSs and electrolyte. Moreover, the In<sub>2</sub>O<sub>3</sub> NSs-based PEC VBUV PDs reveal good wavelength selectivity for visible-blind detection. The UV-visible rejection ratio  $R_{254}/R_{455}$  is 1567, which is  $\approx 30$  times higher than that of In<sub>2</sub>O<sub>3</sub> NPs and exceeds all recently reported PEC VBUV PDs. Moreover, ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs show good multicycle and long-term stability for 254 nm irradiation. Further, In2O3 NSs PEC PDs exhibit good underwater imaging capability.

#### 2. Results and Discussion

#### 2.1. Characterizations of $In_2O_3$ NSs

 $In_2O_3$  NSs were synthesized by following an earlier report<sup>[28]</sup> (more details in Supporting Information). Figure 1a shows the X-ray diffraction (XRD) patterns of the synthesized samples. All the peaks are in accordance with the standard card (PDF

No.06-0416) and can be indexed to the characteristic peaks of the cubic In<sub>2</sub>O<sub>3</sub> sample.<sup>[28]</sup> No other impure diffraction peaks are observed, indicating good purity. The morphology of the synthesized In<sub>2</sub>O<sub>3</sub> samples was observed by transmission electron microscopy (TEM). The synthesized In<sub>2</sub>O<sub>3</sub> samples show a typical nanosheet morphology with lateral sizes in the range of 20-40 nm (Figure 1b and Figure S1a, Supporting Information). Figure 1b inset is the high-resolution TEM (HRTEM) image of In<sub>2</sub>O<sub>3</sub> NSs, which further demonstrates its good crystallinity. The lattice spacings of the fringe pattern are 0.29 and 0.41 nm, corresponding to the (222) and (211) planes of the cubic In<sub>2</sub>O<sub>3</sub> crystal, respectively, which are consistent with an earlier report.<sup>[28]</sup> All Raman peaks correspond to characteristic peaks of In<sub>2</sub>O<sub>3</sub> (Figure S1b, Supporting Information), further demonstrating the successful preparation of the In2O3 sample. Atomic force microscopy (AFM) confirms that the thickness of In2O3 NSs is ≈3.5–4.5 nm, as shown in Figure 1c, demonstrating their ultrathin feature. The morphology of the commercial In<sub>2</sub>O<sub>3</sub> NPs was observed by scanning electron microscopy (SEM), and the sizes of the In<sub>2</sub>O<sub>3</sub> NPs are in the range of 40-60 nm, as shown in Figure S2, Supporting Information. The light absorption properties of In<sub>2</sub>O<sub>3</sub> NSs and NPs were measured by UV-vis diffuse reflectance spectroscopy (Figure 1d and Figure S3, Supporting Information). Ultrathin In2O3 NSs have strong UV absorption, indicating their visible-blind photoresponse capability. Compared with commercial In<sub>2</sub>O<sub>3</sub> NPs, the light absorption of ultrathin In2O3 NSs shifts to the visible-blind region, as shown in Figure 1d and Figure S3a, Supporting Information.

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The blueshift of light absorption is attributed to the quantum confinement effect of ultrathin In<sub>2</sub>O<sub>3</sub> NSs.<sup>[27]</sup> Due to the optical bandgap of In<sub>2</sub>O<sub>3</sub> is under debate (direct or indirect bandgap), both direct and indirect optical bandgaps of In<sub>2</sub>O<sub>3</sub> NSs are calculated by the Tauc curve using the equation of  $(\alpha h v)^{1/n} = B \times$  $(hv - E_g)$ , where n values are 0.5 and 2 for direct bandgap and indirect bandgap semiconductor,  $\alpha$ , hv, B, and  $E_{\alpha}$  refer to the light absorption coefficient, energy of the photon, constant, and bandgap of ultrathin In<sub>2</sub>O<sub>3</sub> NSs, respectively. The direct optical bandgap of ultrathin In<sub>2</sub>O<sub>3</sub> NSs is 3.67 eV calculated by Tauc curves, as shown in Figure 1d inset, which is larger than that of commercial In<sub>2</sub>O<sub>3</sub> NPs in Figure S3b, Supporting Information. Meanwhile, the indirect optical bandgap of ultrathin In<sub>2</sub>O<sub>3</sub> NSs is also larger than that of commercial In<sub>2</sub>O<sub>3</sub> NPs in Figure S3b,c, Supporting Information, further demonstrating their potential in visible-blind detection.

#### 2.2. Photoresponse of Ultrathin In2O3 NSs PEC PDs

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To investigate the photoresponse of ultrathin In<sub>2</sub>O<sub>3</sub> NSs photoanodes (more details in the Experimental Section), a standard three-electrode system was designed, including working electrode (In<sub>2</sub>O<sub>3</sub> NSs photoanodes, Figure S4, Supporting Information), reference electrode (Ag/AgCl), and counter electrode (Pt wire). The photoresponse of In<sub>2</sub>O<sub>3</sub> NSs PEC PDs was recorded by an electrochemical workstation (Scheme 1). There is a built-in electric field pointing from n-type In<sub>2</sub>O<sub>3</sub> to electrolytes after the In<sub>2</sub>O<sub>3</sub> NSs photoanodes immersion in the solution.<sup>[26]</sup> The photogenerated electrons flow from In<sub>2</sub>O<sub>3</sub> to FTO and reach to counter electrode via external circuit. Meanwhile, photogenerated holes transfer from In<sub>2</sub>O<sub>3</sub> to the In<sub>2</sub>O<sub>3</sub>/electrolyte interface and react with hydroxyl (OH<sup>-</sup>), producing hydroxyl radical (OH•). The OH• spreads to the counter electrode and reacts with electrons, completing the photogenerated carriers transport cycle. Therefore, In2O3 NSs PEC PDs could have self-powered photodetection capability. The self-powered PDs can transfer the light signal into an electrical signal without external power. Except for PEC PDs, the conventional solid-state PDs based on p-n or Schotty junctions also show self-powered functions. Unlike PEC PDs involving physical and chemical processes, the conventional solid-state self-powered PDs only involve physical processes, where the photogenerated carriers (electron and hole) are separated due to the built-in electric field, showing a photovoltaic effect.

Ultrathin In2O3 NSs show strong optical absorption in the UV region (Figure 1d), therefore, we investigated the photoresponse behavior of ultrathin In2O3 NSs PEC PDs irradiated by 254 nm with different bias voltages and light power intensities (Table S1, Supporting Information), as shown in Figure 2a. Under 254 nm irradiation, ultrathin In2O3 NSs PEC PDs show a typical on-off switching response. Generally, photocurrent density  $(J_{ph})$ , responsivity (R), and specific detectivity ( $D^*$ ) are key parameters for quantitatively evaluating the performance of PDs, which can be calculated by the following formulas: 1)  $J_{ph} = J_{light} - J_{dark}$ , 2)  $R = J_{\rm ph}/P$ , and 3)  $D^* = R \times S^{0.5}/(2 \times q \times J_{\rm dark} \times S + 4 \times k_{\rm b} \times T/2)$  $R_0$ )<sup>0.5</sup>, where  $J_{\text{light}}$  and  $J_{\text{dark}}$  represent the current density with and without irradiation, S, q, P, and R<sub>0</sub> correspond to the effective area of  $In_2O_3$  NSs on FTO (1 × 1 cm<sup>2</sup>), the quantity of electron charge ( $1.602 \times 10^{-19}$  C), the light power intensity, and the resistance, respectively. The D\* values are calculated by considering both shot noise and thermal noise,<sup>[29,30]</sup> as shown in Tables S4 and S5, Supporting Information. At a fixed bias voltage, J<sub>ph</sub> gradually increases with the increment of P due to more photogenerated carriers at stronger P. For example, J<sub>ph</sub> increases from 6.86 to 10.16  $\mu$ A cm<sup>-2</sup> as the *P* increases from level I to V at a bias voltage of 0.4 V (more details in Table S2, Supporting Information). At a fixed *P*, the  $J_{\rm ph}$  rapidly increases from 0 to 0.4 V, as shown in Table S2, Supporting Information. For example,  $J_{ph}$  is 0.33, 1.53, and 6.86  $\mu A~cm^{-2}$  for 0, 0.2, and 0.4 V at 0.04  $mW~cm^{-2}$ (level I), respectively. The  $J_{\rm ph}$  at 0.4 V is 21-fold higher than that of 0 V (level I). Although ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs show a self-driven photodetection capability,  $J_{ph}$  is relatively small at 0 V (Figure S5a, Supporting Information). The  $J_{\rm ph}$  approaches saturation at a higher bias voltage of 0.6 V, as shown in Figure S5b, Supporting Information, attributing to saturation photogeneration carriers transport at higher bias voltage. Therefore, we will focus on the performance of ultrathin In2O3 NSs PEC PDs at 0.4 V. Furthermore, ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs show good reproducibility for different devices with small deviations, as shown in Tables S2-S4, Supporting Information. As shown in Figure 2b,c, ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs show an outstanding photoresponse to 254 nm with an ultrahigh *R* of 172.36 mA  $W^{-1}$ and a remarkable  $D^*$  of 4.43  $\times$  10<sup>11</sup> Jones under 254 nm irradiation at 0.4 V, respectively, which surpass all recently



Scheme 1. Schematic diagram of evaluating ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs.







**Figure 2.** Photoresponse of ultrathin  $In_2O_3$  NSs PEC PDs under 254 nm irradiation. a) *J*-*t* curves at various bias voltages and light power intensities (levels I, II, III, IV, and V correspond to 0.04, 0.08, 0.12, 0.14, and 0.22 mW cm<sup>-2</sup>, respectively). b) Corresponding *R*. c) Corresponding *D*\*. d) *J*-*t* curves of ultrathin  $In_2O_3$  NSs PEC PDs in various concentrations of electrolyte, including 0.01, 0.05, 0.1, 0.2, and 0.5 M Na<sub>2</sub>SO<sub>4</sub>. e) Corresponding *J*<sub>ph</sub> in various concentrations of Na<sub>2</sub>SO<sub>4</sub> solution. f) EIS of  $In_2O_3$  NSs PEC PDs in different concentrations of electrolyte.

reported ultrathin nanomaterial-based aqueous-type PEC UV PDs<sup>[31-49]</sup> and are comparable with the record-high performance of recently reported PEC UV PDs,<sup>[17]</sup> as shown in Table S9, Supporting Information. The ultrahigh UV detection capability and the operation in weak alkaline electrolyte indicate that ultrathin In<sub>2</sub>O<sub>3</sub> NSs hold great prospects for underwater UV communication<sup>[12]</sup> (more detailed comparison in Table S6, Supporting Information). We will focus on investigating optoelectronic properties of In<sub>2</sub>O<sub>3</sub> NSs PEC PDs using seawater as the electrolyte in the future.

For PEC PDs, the electrolyte is another parameter to manipulate the photoresponse. In this study, Na<sub>2</sub>SO<sub>4</sub> is chosen as the electrolyte because In<sub>2</sub>O<sub>3</sub> is unstable in alkali and acid solutions. The photoresponse of ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs was measured in various concentrations of Na<sub>2</sub>SO<sub>4</sub> solution ranging from 0.01 to 0.5 м. Ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs show similar on-ff switching behaviors under 254 nm irradiation in different concentrations of electrolyte, shown in Figure 2d. With increasing  $Na_2SO_4$  concentration in the range of 0.01 - 0.5 M,  $J_{ph}$  gradually increases from 3.31 to 6.86  $\mu$ A cm<sup>-2</sup> at level I (Figure 2e and Table S7, Supporting Information). To further explore the mechanism of the electrolyte concentration-dependent photoresponse, we measured the electrochemical impedance spectroscopy (EIS) of ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs in different concentrations of Na<sub>2</sub>SO<sub>4</sub> solutions, shown in Figure 2f and Table S8, Supporting Information. The  $R_{ct}$  and series resistance  $(R_s)$ reduce gradually with increasing Na2SO4 concentration, indicating that a higher concentration of Na<sub>2</sub>SO<sub>4</sub> accelerates charge transport and facilitates photogenerated hole transfer between In<sub>2</sub>O<sub>3</sub> NSs and electrolytes, leading to a higher photoresponse.

#### 2.3. Spectral Photoresponse of Ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs

To understand the spectral photoresponse behavior of In<sub>2</sub>O<sub>3</sub> NSs, the continuous spectral photoresponse (240-500 nm) was measured, as shown in Figure S6, Supporting Information. The photoresponse dramatically reduces as the wavelength of irradiation light increases, which is consistent with the light absorption properties. The PEC PDs show negligible photoresponse as the wavelength of irradiation light is longer than 400 nm, indicating their VBUV detection capability. To more accurately evaluate the spectral photoresponse behavior of In<sub>2</sub>O<sub>3</sub> NSs, four different wavelengths of light (254, 365, 455, and 525 nm) were used to irradiate ultrathin In2O3 NSs PEC PDs in 0.5 м Na<sub>2</sub>SO<sub>4</sub> at a fixed voltage of 0.4 V, as shown in Figure 3a. Obvious on-off switching signals are exhibited in the UV region (254 and 365 nm), and the  $J_{\rm ph}$  signal rapidly declines in the visible region (455 and 525 nm). Compared with ultrathin In<sub>2</sub>O<sub>3</sub> NSs, In2O3 NPs show a wide photoresponse region from UV to visible, as shown in Figure S7, Supporting Information. The spectral selectivity agrees well with the UV-vis absorption spectrum of ultrathin In<sub>2</sub>O<sub>3</sub> NSs and commercial In<sub>2</sub>O<sub>3</sub> NPs. The spectral selectivity is quantitatively evaluated by UV/visible rejection ratio  $(R_{254 \text{ nm}}/R_{455 \text{ nm}})$ , which are 1567 and 45.9 for ultrathin In2O3 NSs and In2O3 NPs, respectively, demonstrating the boosted spectral selectivity of ultrathin In<sub>2</sub>O<sub>3</sub> NS. The spectral selectivity of ultrathin In2O3 NS is approximately nine and 30 times higher than those of In2O3 microrods<sup>[26]</sup> and commercial In<sub>2</sub>O<sub>3</sub> NPs, respectively. The improved spectral selectivity is attributed to the wider bandgap of ultrathin In<sub>2</sub>O<sub>3</sub> NSs originating from the quantum confinement effect.<sup>[27]</sup> The spectral

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**Figure 3.** Spectral photoresponse of ultrathin  $In_2O_3$  NSs PEC PDs. a) *J*-*t* curves of  $In_2O_3$  NSs PEC PDs irradiated by 254, 365, 455, and 525 nm at a bias voltage of 0.4 V in 0.5 M  $Na_2SO_4$  with a light power intensity of level I; b) corresponding *R* of  $In_2O_3$  NSs PEC PDs irradiated by 254, 365, 455, and 525 nm; c) comparison of rejection ratios and *R* of recently reported aqueous-type PEC UV PDs. d) EIS of  $In_2O_3$  NSs and NPs PEC PDs in 0.5 M  $Na_2SO_4$ .

selectivity and *R* of ultrathin  $In_2O_3$  NSs PEC VBUV PDs outperform most reported aqueous-type PEC VBUV PDs,<sup>[26,44–48]</sup> as shown in Figure 3c, demonstrating their great potential in VBUV detection.

Ultrathin In<sub>2</sub>O<sub>3</sub> NSs exhibit a higher photoresponse than In<sub>2</sub>O<sub>3</sub> NPs in the UV region, which contradicts light absorption (Figure S3a, Supporting Information). To clarify this phenomenon, EIS of ultrathin In<sub>2</sub>O<sub>3</sub> NSs and commercial In<sub>2</sub>O<sub>3</sub> NPs PEC PDs in 0.5  $\bowtie$  Na<sub>2</sub>SO<sub>4</sub> solutions were recorded under 254 nm irradiation, as shown in Figure 3d. Ultrathin In<sub>2</sub>O<sub>3</sub> NSs and commercial In<sub>2</sub>O<sub>3</sub> NPs show almost same the fitting  $R_{\rm s}$  in Table S8, Supporting Information, demonstrating their similar charge transport capability. The fitting  $R_{\rm ct}$  of ultrathin In<sub>2</sub>O<sub>3</sub> NSs is 5.797  $\Omega$ , which is 63% that of In<sub>2</sub>O<sub>3</sub> NPs, demonstrating a stronger interfacial charge transfer capability at the ultrathin In<sub>2</sub>O<sub>3</sub> NSs/electrolyte interface. The EIS results indicate that the higher UV photoresponse of ultrathin In<sub>2</sub>O<sub>3</sub> NSs is due to easier interfacial charge transfer rather than stronger UV absorption.

# 2.4. Photoresponse Time and Stability of Ultrathin $\rm In_2O_3$ NSs PEC PDs

The response speed and stability of ultrathin  $In_2O_3$  NSs PEC PDs were investigated by 254 nm at 0.4 V. **Figure** 4a shows an enlarged the photo-switching cycle. The rise time ( $T_r$ ) represents the time that the current rises from 10% to 90% of the maximum current, and the decay time ( $T_d$ ) is the time of the maximum current reducing from 90% to 10%. The rise time

 $(T_r)$  and decay time  $(T_d)$  of ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs are 0.8 s and 2.2 s, respectively, which are comparable to those of some aqueous-type PEC UV PDs.<sup>[26,47,49]</sup> The slower decay time is due to the persistent photoconductivity effect, which is caused by large quantity of oxygen vacancy defects and high density of trap states in metal oxide semiconductors.<sup>[50,51]</sup> The multicycle and long-term stability are key parameters for PDs in practical applications. Figure 4b shows the multicycle and long-term stability tests of In<sub>2</sub>O<sub>3</sub> NSs PEC PDs in 0.5 м Na<sub>2</sub>SO<sub>4</sub>, which were continuously tested for 2000 s. For the fresh sample (storage for 1 month),  $J_{\rm ph}$  decreases from 9.6 (7.4) to 8.84 (7.2)  $\mu A \ \rm cm^{-2}$ after 2000 s, demonstrating good stability under 254 nm irradiation. The decrease in  $J_{\rm ph}$  may be due to decomposition of nanocrystals or pilling-off from the substrate. Ultrathin In<sub>2</sub>O<sub>3</sub> PEC PDs exhibit good multicycle and long-term stability, which are superior to most aqueous-type ultrathin nanomaterial-based PEC UV PDs.<sup>[31-43]</sup>

#### 2.5. Imaging Capability of Ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs

To further verify the capability of our devices for underwater imaging applications, the ultrathin  $In_2O_3$  NSs PEC PDs are employed in a single-pixel imaging system. As shown in **Figure 5**a, the imaging system consists of a hollow plate with the school badge (Northeast Forestry University), a laser light source (375 nm, 0.06 mW cm<sup>-2</sup>), a source meter (Keithley 6482), an oscilloscope, and a computer. The object is mounted on a computer-controlled X-Y platform so that it can move continuously and stably in both horizontal and vertical directions,

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**Figure 4.** Photoresponse time and stability of ultrathin  $In_2O_3$  NSs PEC PDs. a) Amplified photo-switching cycle irradiated by 254 nm (Level III) in 0.5 M  $Na_2SO_4$ . b) Stability measurements of fresh samples and storage for 1 month for 2000 s (light on 5 s and light off 5 s).

and the current signals are collected by the source meter at the same time. A  $103 \times 103$  pixels image is obtained under the bias of 0.4 V, as shown in Figure 5b. The clear and accurate pattern of the school badge proves the underwater imaging capability of the device. Figure 5c shows the profile of the red line in Figure 5b, and the rapid increases and decreases of signal intensity prove the device's reliability when applied to underwater imaging. The imaging process can also be achieved in a self-powered mode. As shown in Figure S8, Supporting Information, limited by ambient noise and instrument accuracy, the

image obtained in self-powered mode is slightly worse but can be distinguished. These imaging results demonstrate the potential application capabilities of ultrathin  $In_2O_3$  NSs PEC PDs.

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# 3. Conclusion

In summary, we first demonstrated that individual ultrathin  $In_2O_3$  NSs have great potential in high-performance PEC VBUV PDs with high responsivity and excellent spectral



Figure 5. Imaging Capability of ultrathin  $In_2O_3$  NSs PEC PDs. a) Schematic diagram of the imaging system. b) Imaging under a bias of 0.4 V. c) Detected signal along the red line in (b).



selectivity. Ultrathin In2O3 NSs PEC PDs show excellent UV photodetection with an ultrahigh R of 172.36 mA  $W^{-1}$  and a remarkable  $D^*$  of  $4.43 \times 10^{11}$  Jones under 254 nm irradiation. The outstanding UV photoresponse originates from the smaller charge transfer resistance at the ultrathin In<sub>2</sub>O<sub>3</sub> NSs/electrolyte interface. Furthermore, the light absorption of ultrathin In<sub>2</sub>O<sub>3</sub> NSs takes a blueshift due to the quantum confinement effect, granting ultrathin In2O3 NSs PEC PDs good spectral selectivity for visible-blind detection. The UV-visible rejection ratio of In<sub>2</sub>O<sub>3</sub> NSs PEC PDs is 1567, which exceeds all recently reported PEC VBUV PDs. Moreover, ultrathin In<sub>2</sub>O<sub>3</sub> NSs PEC PDs show good multicycle and long-term stability under 254 nm irradiation. Furthermore, In<sub>2</sub>O<sub>3</sub> NSs PEC PDs show good underwater imaging capability under UV irradiation. Our results demonstrate that ultrathin In<sub>2</sub>O<sub>3</sub> NSs are good candidates for highperformance VBUV PDs.

### 4. Experimental Section

Photoresponse Activity: In brief, In2O3 (2 mg) NSs/NPs were dispersed into 1 mL of PVDF/DMF (2 mg/10 mL) by sonicating for 60 min. The mixture was then directly dropped onto the conductive side of FTO glass, and the In<sub>2</sub>O<sub>3</sub>-coated FTO (effective area of In<sub>2</sub>O<sub>3</sub> NSs on FTO was  $1 \times 1$  cm<sup>2</sup>) was dried in a vacuum drying oven at 80 °C for 12 h. The electrical characterization was carried out in a three-electrode system using a CHI660E electrochemical workstation (Chenhua Instrument Company, China). In<sub>2</sub>O<sub>3</sub> NSs/NPs-coated FTO glass, Pt wire, and Ag/AgCl electrodes were used as the working electrode, counter electrode, and reference electrode, respectively. The photodetection performance was investigated in different concentrations of  $Na_2SO_4$  (0.01, 0.05, 0.1, 0.2, and 0.5 M) with different wavelengths of light (254, 365, 455, and 525 nm). The electrochemical impedance spectrum (EIS) was determined in a frequency range of 0.01 Hz-10<sup>5</sup> Hz with a perturbation amplitude of 0.005 V. Measurements of continuous spectral responsivity were performed using a Zolix DR800-CUST and a CHI660E electrochemical workstation. A monochromatic light with a continuously tunable wavelength (200-500 nm) illuminated the sample surface, and the generated photocurrents from the In2O3 NSs PEC PDs were collected by the electrochemical workstation. After testing, the light intensity of monochromatic light was measured with the optical power meter (ThorLabs, PM120VA). The imaging system consisted of a hollow plate with the school badge (Northeast Forestry University), a laser light source (375 nm, 0.06 mW cm<sup>-2</sup>), a source meter (Keithley 6482), an oscilloscope, and a computer. The school badge was mounted on a computer-controlled X-Y platform. A  $103 \times 103$  pixels image was obtained under the bias of 0 and 0.4 V.

Statistical Analysis: Statistical analyses were conducted using OriginPro 2018 (OriginLab Software, Northampton, Massachusetts, USA). Data from photocurrent density, responsivity, and specific detectivity of  $In_2O_3$  NSs PEC PDs irradiated by 254 nm with different power intensities were calculated and processed as mean  $\pm$  standard deviation (SD). The error bars were based on the SD of the mean.

# **Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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# **Conflict of Interest**

The authors declare no conflict of interest.

## Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

### **Keywords**

In2O3, nanosheets, photodetectors, photoelectrochemical, visible-blind

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