Visible Light-Assisted High-Performance Mid-Infrared Photodetectors Based on Single InAs Nanowire

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Supporting Information

ABSTRACT: One-dimensional InAs nanowires (NWs) have been widely researched in recent years. Features of high mobility and narrow bandgap reveal its great potential of optoelectronic applications. However, most reported work about InAs NW-based photodetectors is limited to the visible waveband. Although some work shows certain response for near-infrared light, the problems of large dark current and small light on/off ratio are unsolved, thus significantly restricting the detectivity. Here in this work, a novel “visible light-assisted dark-current suppressing method” is proposed for the first time to reduce the dark current and enhance the infrared photodetection of single InAs NW photodetectors. This method effectively increases the barrier height of the metal–semiconductor contact, thus significantly making the device a metal–semiconductor–metal (MSM) photodiode. These MSM photodiodes demonstrate broadband detection from less than 1 μm to more than 3 μm and a fast response of tens of microseconds. A high detectivity of ~10^12 Jones has been achieved for the wavelength of 2000 nm at a low bias voltage of 0.1 V with corresponding responsivity of as much as 40 A/W. Even for the incident wavelength of 3113 nm, a detectivity of ~10^10 Jones and a responsivity of 0.6 A/W have been obtained. Our work has achieved an extended detection waveband for single InAs NW photodetector from visible and near-infrared to mid-infrared. The excellent performance for infrared detection demonstrated the great potential of narrow bandgap NWs for future infrared optoelectronic applications.

KEYWORDS: Single InAs nanowire, mid-infrared photodetectors, MSM photodiodes

Indium arsenide nanowires (InAs NWs), have been widely researched in recent years together with other III–V semiconductor NWs like InP,1–3 GaN,4–6 InSb,7–9 and so forth. With a high mobility and direct narrow bandgap (approximately 0.35 eV), InAs NWs seem to be more appropriate for applications of high-speed electronic components and broad-spectrum detection media.7,10–17 Photodetectors based on single InAs NW and NW arrays have been successfully fabricated and exhibit a good photoresponse.10,11,13,18–21 Research has identified two different photodetection mechanisms for single InAs NW photodetector, which are positive photoresponse (PPR)11,13 and negative photoresponse (NPR).22–24 The difference between these two mechanisms is that the latter is a phenomenon induced by surface states similar to a photogating layer (PGL) trapping hot electrons and seems to work only when the energy of incident photon is much higher than the bandgap of InAs.23 For NPR, an ultrahigh photoconductive gain of ~10^9 and a response time of less than 5 ms have been achieved.22,23 For PPR, the responsivity and detectivity reach 5.3 × 10^7 A/W and

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and trapped as an NPR process, whereas the trapped- 
light, hot electrons are excited into the PGL before attempting infrared detection. Because of the illumina-
tion of 450 nm light. (d) Working principle for infrared light detection under a low bias voltage.

For most NW-based photodetectors designed so far, the net photocurrent is not so considerable unless a junction area was formed, such as Schottky junction, p–n junction, and heterojunction. Recently a new kind of InP NW photodetector enhanced by ferroelectric field was demonstrated. The dark current of these photodetectors is very low due to the high ferroelectric field depletion, leading to ultra-high detectivity. As for simple single InAs NW photodetectors, in addition to weak optical absorption and short carrier lifetime, the large dark current and surface defects scattering noise could be two main factors for the low photoresponse to infrared light. Additionally, the well-known surface electron accumulation layer has a large impact on carrier transport, giving rise to a lower-infrared detectivity. To achieve high sensitivity for detection of infrared light, it is of great importance to reduce the dark current and improve the light on/off ratio. Here we demonstrate a method to realize a broadband detection of single InAs NW from 830 to 3133 nm. Different from other conventional detection methods, a precursor irradiation by 450 nm source whose photon energy is much higher than the energy bandgap of InAs is first used before attempting infrared detection. Because of the illumination of precursor light, hot electrons are excited into the PGL and trapped as an NPR process, whereas the trapped-electrons-releasing process is partly blocked at low temperature. The electrons accumulated on the NW surface result in space charge and strongly enhance the Schottky barrier. The two back-to-back Schottky junctions combined with InAs NW form a metal–semiconductor–metal (MSM) photodiode. Taking advantage of this method, a high detectivity of 2 × 10^12 Jones and a response time of 80 μs have been achieved for the wavelength of 2000 nm at a low bias voltage of 0.1 V.

Figure 1 schematically depicts the working principle. The device schematic is shown at the top-right corner. Before the precursor light illumination, free electrons are driven by the electric field in the NW core and form the dark current. The contact barrier between the InAs NW and metal (Cr/Au) is low or even ohmic (see Figure 1c, left).

When the NW is exposed to the 450 nm laser, the photogating effect occurs (see Figure 1a). The NW surface states induced by inhomogeneous composition or native oxide layer act as the PGL that traps the photoexcited electrons, while photogenerated holes recombine with the free electrons in the core, leading to a decreasing current. After the laser is switched off, trapped electrons are released to the core through a thermal process. What is noteworthy is that the recovery process can be partly blocked if the temperature is low enough. The electrons unable to be released stay in the PGL and repel near-surface electrons, which results in space charge region and band bending of the surface (see Figure 1b). The surface band bending together with Fermi level pinning strongly enhances the barrier between the two metal electrodes and InAs NW (see Figure 1c).

This back-to-back Schottky barrier significantly allows the formation of an MSM photodiode. For an MSM photodiode, fast response and high-frequency bandwidth are the main performance parameters due to the small stray capacitance. When the device is operating at a low bias voltage, few electrons overcome the high back-to-back barriers and the dark current is very small. When under infrared illumination, photogenerated electron–hole pairs in the junction area at the negative voltage point are quickly separated and collected by the electrodes (see Figure 1d). Because the dark current is quite low, this illumination would induce an obvious light current. According to our measurements, the photocurrent can be several orders of magnitude larger than the dark current for infrared light, and this is nearly impossible for direct detection. On the basis of the above discussion, a visible light-assisted and single InAs NW-based MSM photodiode has been designed for infrared detection.

In this paper, InAs NWs were synthesized by the molecular beam epitaxy (MBE) technique and transferred to Si/SiO2 (300 nm) substrate to fabricate the source/drain (S/D) electrodes (see more details in Methods). The diameter of the selected NW is about 80 nm, and the final NW photodetector has a channel of 2.2 μm (see Supporting Information Figure S1a). InAs NWs are known to have a native oxide layer, which could act as the PGL. During the device fabrication process, only the oxide layer at metal–semiconductor contact area was removed before metallization to ensure good contact. Different
than the InAs NWs synthesized by chemical vapor deposition method studied by Guo, our NWs show a relatively smooth surface without a highly defected shell (see Supporting Information Figure S1b). Nevertheless, the NW surface is still structurally and compositionally inhomogeneous, as marked by a red dashed rectangle in Figure S1b, which objectively permits the existence of PGL. We first measured the $I_{ds}-V_{ds}$ characteristics of our NW photodetector under illumination of different wavelengths in ambient environment (see Supporting Information Figure S2a). The photoresponse of our transistor has a strong relation to the incident wavelength. Consistent with other reported work, our NWs show NPR behavior when the photon energy is much higher than the energy bandgap, for example, at $\lambda = 520$ nm, whereas the PPR occurs at the incident wavelength of 940 nm (which has a lower photon energy). This conclusion is also verified by the current measurements in light-modulation mode at a low bias voltage (Figure S2b). We have fabricated tens of devices and although not all the NWs show obvious PPR in NIR waveband, almost all the devices show clear NPR in the visible waveband, especially at $\lambda = 450$ nm. In an ambient environment, a higher incident power is required to achieve a small light current, while at 77 K it would be quite easy (see Figure S2c,d), because the thermal-assisted trapped-electrons-releasing process is seriously blocked. To cause more electrons to be trapped by PGL, a 450 nm laser ($\sim 0.3$ mW/mm$^2$) is used at 77 K in the following studies.

To demonstrate the significance of the design of the visible light-assisted single InAs NW MSM photodiode, we compare the photoresponse of our NW photodetector at $\lambda = 2000$ nm before and after 450 nm laser illumination (the power density is $\sim 0.3$ mW/mm$^2$) at 77 K. As shown in Figure 2a, before illumination, the linear $I_{ds}-V_{ds}$ characteristics indicates a good contact between the metal and InAs NW, while the photocurrent for 2000 nm illumination (power density is $\sim 1.7$ mW/mm$^2$) is too weak to be distinguished. Note that the current is on the level of microamperes ($V_{ds} = 1$ V), which is common for single InAs photoconductive detectors. After the assistant illumination from the 450 nm laser, a clear $I_{ds}-V_{ds}$ characteristic of MSM photodiode is performed (refer to the inset in Figure 2b). When the NW is operated at a low bias (<1 V) because of the back-to-back Schottky barrier the dark current remains at a very low level. Because of the high and thick Schottky junctions, an illumination of 2000 nm at this time induces an obvious photocurrent, which is approximately three orders larger than the dark current (Figure 2b). It is known that the main voltage drop occurs at the reverse biased Schottky barrier when the bias voltage is low. Hence, electron–hole pair generation and separation at the negative bias region is the main source of the large photocurrent (see Figure 1d). Both the photocurrent and dark current increase when the bias voltage increases. When the bias voltage is high enough ($\sim 1.5$ V in Figure 2b), that is, when the bias voltage reaches the breakdown voltage, photocurrent and dark current are almost coincident, and as in the situation before the illumination of 450 nm they are difficult to be distinguished. This situation is mainly attributed to the large number of holes injected into the NW from the positive bias point, which

Figure 2. Comparison of the device performances before and after the illumination of 450 nm light at 77 K. (a) Linear $I_{ds}-V_{ds}$ characteristics in dark and under illumination of 2000 nm before assistant light illumination. (b) Logarithmic $I_{ds}-V_{ds}$ characteristics in dark and under illumination of 2000 nm after assistant light illumination. Inset: the MSM feature presented by linear $I_{ds}-V_{ds}$ characteristics in dark. (c) Photoresponse properties of the MSM photodiode under a 0.5 Hz-chopped 2000 nm laser illumination, measured at $V_{ds} = 0.1$ V. (d) Comparison of the responsivity and detectivity for $\lambda = 2000$ nm before and after 450 nm light illumination, measured at $V_{ds} = 0.1$ V. All the power densities of the 2000 nm laser used here were approximately 1.7 mW/mm$^2$.
dominates the current and makes the photocurrent too weak to be evident. Note that the $I_{ds} - V_{ds}$ curve in the dark conditions does not go through the origin. This may be attributed to the capacitance difference of the two junctions. To present a clearer photoresponse at 2000 nm, photoconductance modulation was performed at a low bias voltage of 0.1 V. As shown in Figure 2c, the dark current is only several picoamperes, while the photocurrent is about 16 nA with a fast response to a 0.5 Hz chopped wave. Response speed is vital for a photodetector and a response time of 80 μs was obtained through a high-resolution measurement performed later (see Figure 4c), which is faster than that of previously reported common InAs NW-based photodetectors (summarized in Figure 4f). In addition to response time, responsivity ($R$) and specific detectivity ($D^*$) are two key performance parameters. The responsivity of a photodetector can be expressed in units of amperes per watt of incident radiant power, defined as $R = I_{ph}/(PA)$, where $I_{ph}$ is the net photocurrent, $P$ is the light power density, and $A$ is the effective area of the detector. $D^*$ is a figure of merit used to characterize performance, given as $D^* = (AB)^{1/2}/NEP$, where NEP is the noise equivalent power and $B$ is the bandwidth. Considering that the shot noise from dark current is the major factor limiting detectivity, the specific detectivity can be simply expressed as $D^* = RA^{1/2}/(2eI_{dark})^{1/2}$, where $e$ is the...
simplification of the device (see Supporting Information Figure S3). However, due to the high back-to-back barrier, the dark current which makes the noise and background di

To demonstrate the validity of the mechanism discussed above, the importance of the PGL needs to be further confirmed. Previous study showed that the adsorption of gas molecules such as O$_2$ could also promote the trapping of electrons. Surface electrons trapped by adsorbed chemical molecules can also modulate the Schottky barrier height like a floating gate. Therefore, it is necessary to prove that our visible light-assisted method still functions without adsorption of gas molecules. Thermal desorption is an effective way to remove the surface adsorbed gas molecules. In the first step, the device was heated from 300 to 460 K in a vacuum environment ($\sim 10^{-5}$ Torr) with the turbo pump operating continuously. With increasing temperature, the drain current is obviously increased (see Supporting Information Figure S4). Since our NWs have been previously annealed, this increasing current is mainly attributed to the gas desorption occurring on NWs surface. Finally, the device was maintained at 460 K for more than half an hour. Even at 460 K, the NPR is still readily apparent (see Figure 3a) and repeatable. We compared the NPR at 460 and 360 K (see Figure 3b) and found that at 460 K the current recovery process is faster. The parameter $\tau$ from the fitting curve through the formula $y = y_0 + A \exp(-(x-x_0)/\tau)$ directly demonstrates this point. We have tested more than 10 NWs and it was found that although their recovery time varies from several seconds to several minutes or even longer, all the device currents can recover. This provides a direct support for our hypothesis that the NPR process is mainly dominated by the native PGL but not gas molecules. Furthermore, we showed that a lower operating temperature would lead to a longer current recovery process. This thermal-assisted electrons-releasing process has also been demonstrated by other previous work. We subsequently cooled the device from 460 to 77 K in dark conditions and found that the drain current became smaller while without so much change (see dark line in Figure 3a,c). While after repeating the visible light-assisted method by exposure of the device to 450 nm laser illumination (0.3 mW/mm$^2$), the junctions were formed again (see Figure 3c, dark line). This result provides significant evidence for the important role of the PGL itself. However, since there are always many gas molecules surrounding the InAs NWs, even in a high vacuum environment of $10^{-5}$ Torr, the cooling process may still bring in gas adsorption, and this impact cannot be completely ignored, especially for oxygen molecules. Previous research results show the adsorption of oxygen could create surface states in the band gap. This is also consistent with the incomplete releasing of trapped electrons at low temperature. Additionally, electrostatic interaction could gather more O$_2$ in the junction area, further enhancing the barrier heights. We hypothesize that the gas adsorption could work together with the PGL at low temperature, however the latter is indeed the basis of our detection method. At least without the illumination of 450 nm light, the formation of such a large Schottky barrier is nearly impossible.

For the trapped electrons in the PGL, a back gate voltage ($V_g$) sweep or a negative gate voltage pulse would pull some of them back to the core. Figure 3c shows the $I_{ds}-V_{ds}$ characteristics measured at 77 K under three different conditions, dark (without any illumination, dark line), after 450 nm laser irradiation (formation of the MSM photodiode, blue line), and then after a $V_g$ sweep from $-40$ to 40 V (red line), respectively. Obviously the drain current of “After $V_g$ sweep” was much larger than that of “After 450 nm”. It means by a $V_g$ sweep, the free carrier concentration in the NW channel became larger and the high Schottky barriers were reduced. However, the drain current is still less than that of initial state, indicating an incomplete releasing of surface trapped electrons. Nevertheless, the formation of the MSM photodiode is repeatable. As shown in Figure 3d, under a chopped illumination of 450 nm ($\sim 0.3$ mW/mm$^2$, without a fixed frequency), the current gradually decreased to a low level of several picoamperes at $V_{ds} = 0.1$ V, indicating that a large number of electrons are trapped and the two high Schottky barriers are formed again. It is interesting to note that when the 450 nm light was switched off for the first time, the current dropped instead of increasing. This is because after a certain illumination duration, the intrinsic excitation in the junction area and NW channel work together, although many electrons are trapped, which reduces the current. PPR of the following chopped cycles also supports this point.

To further understand the source of the photocurrent, we compared the $I_{ds}-V_{ds}$ characteristics of our MSM photodiode in dark conditions and under 450 nm illumination. Here, “in dark conditions” means after the exposure of 450 nm light, and the MSM photodiode has been already formed. As shown in Figure 3e, we concluded that the light current of 450 nm is somewhat larger than the dark current (see points marked by red color in Figure 3e) when the device is operated at a low positive bias voltage (such as 0.1 V). However, when the bias voltage is near 1.6 V, the light current is clearly lower than the dark current, causing a positive shifting of the breakdown voltage. In order to substantiate this result, a modulated light source of 450 nm was used at the different bias voltages of 0.2 and 1.5 V, respectively, with results shown in Figure 3f. At $V_{ds} = 1.5$ V, an NPR was evident. This indicates that at 77 K, even after the formation of the MSM photodiode by a 450 nm laser exposure, there are still some electrons that could be excited to the PGL by 450 nm laser thus further modulating the barrier height but able to be released when the laser is switched off. This could be attributed to shallow surface defect energy levels. While at $V_{ds} = 0.2$ V, due to the two high Schottky barriers, photogenerated electron–hole pairs in the junction area dominate the photocurrent, leading to a remarkable positive responsesignal ($\sim 60$ pA). This is consistent with the current drop when the laser is switched off for the first time as shown in Figure 3d. To further confirm the important role of Schottky junctions, a photocurrent mapping for our InAs NW MSM photodiode was conducted, as shown in Figure 3g. This measurement was performed under 520 nm laser illumination with a power of $\sim 5 \mu$W at 77 K. The laser beam was focused on the device through a 20X objective lens, resulting in a Gaussian-like light spot with a full width half-maximum (fwhm)
The dimension of $\sim 3\ \mu m$. As the channel of the photodiode used here is only about $3\ \mu m$ and the diameter is much smaller than the spot light, the photocurrent mapping is oblate more than virgulate. The two highlighted areas with different sizes near the electrodes suggest a strong photoresponse, while a small area near the positive bias point is dim indicating a much weaker response. The phenomena could be understood as follows. When the laser spot is irradiated on the region near the electrodes, intrinsic excitation and electron−hole separation in the junction dominate the photocurrent. When the laser spot is irradiated on the NW channel, some hot electrons move to the NW surface that would further enhance the barrier height, and some photoexcited electrons and holes in the NW core contribute to the free carrier concentration. However, due to the short lifetime of minority carrier, it is hard to make a contribution to the photocurrent. So the mapping in NW channel is dimmer while bright near the electrodes. The photocurrent was also directly recorded using an oscilloscope, as shown in Figure 3h. The time axis on the oscilloscope can also be regarded as the distance along the NW because the light spot is scanned perpendicularly to the NW and along the NW line-by-line with a uniform velocity. Each time the light spot was scanned across the NW, photocurrent was generated, leading to a sharp peak on oscilloscope. Peak values are fitted using two Gaussian functions (see red line and green line in Figure 3h). Different fwhm and peak values of these two Gaussian functions prove that the Schottky junctions, especially the one near the negative bias point, are the main regions from where the photocurrent originates.

As discussed above, a visible light-assisted and single InAs NW-based photodiode has been demonstrated. It is to be particularly noted that hot electrons are trapped by the PGL and some electrons in the PGL are unable to be released to the core at 77 K. This causes surface band bending and promotes the formation of the two large Schottky barriers. This method is not suitable for visible light detection, because energy in this waveband is still able to excite some electrons to the PGL, even though the MSM photodiode is still sensitive to 450 nm light when working at a low bias voltage. It is worth noting that most devices we fabricated showed either small positive response or no response to infrared light before illumination by precursor light (our visible light-assisted method). Therefore, the MSM photodiode is very suitable for infrared detection. To investigate more detailed performances of our MSM NW photodiode for infrared detection, we first checked the power-dependent photoresponse at a wavelength of 2000 nm. Figure 4a,b shows the responsivity−power relation. When the power density is low (<1.6 mW/mm$^2$), the net photocurrent and responsivity increase with increased power density. However, when the power density is higher (>1.6 mW/mm$^2$), the net photocurrent begins to saturate and the responsivity decreases. Note that the responsivity increases first then decreases, which is different from most reported photodetectors whose responsivity directly decreases with increasing light power density. This may be attributed to the combined function of surface electrons induced electric field and build-in internal electric field in the Schottky junctions. A highest responsivity of 46 A/W is obtained at a power density of 1.6 mW/mm$^2$ for $\lambda = 2000$ nm, and the detectivity reaches $\sim 10^{12}$ Jones. For a photodetector, the response speed is a vital...
parameter that determines the capability of a photodetector to follow a fast-varying optical signal. Figure 4c shows three normalized modulation cycles of 1000 Hz at $V_{ds} = 0.1$ V for the wavelength of 2000 nm. We use the formula $y = y_0 + A \exp(-\frac{(x - x_0)}{\tau})$ to fit the curve, and calculated the response time from 10% to 90%. A rising time of $\sim 80 \mu s$ and a falling time of $\sim 61 \mu s$ have been obtained, respectively. Compared to other InAs NW FET photodetectors, our MSM photodiodes show a much faster photoresponse (summarized in Figure 4f). This fast photoresponse may be attributed to the high-quality NW core and trapped electrons-passivated NW surface (see Supporting Information Figure S5). To determine the wavelength-dependent photoresponse, $I_{dark} - V_{ds}$ characteristics were measured under illuminations of wavelengths from 830 to 3113 nm, as shown in Figure 4d. All the illumination power densities were 0.7 mW/mm². Figure 4e shows the responsivity/detectivity wavelength relation, calculated at a bias voltage of 0.4 V. The responsivity reaches the maximum value near the wavelength of 2000 nm and shows a Gaussian-like distribution with the wavelength. Even for $\lambda = 3113$ nm, R and $D^*$ reach 0.6 A/W and $\sim 10^{10}$ Jones, respectively, satisfying the requirements of most detectors at this wavelength.

We summarize the detection waveband and response time of single InAs NW-based photodetectors reported so far, as shown in Figure 4f. For the first time, our work successfully extended the detection waveband of single InAs NW photodetectors from the visible and near-infrared wavelengths to mid-infrared. Moreover, by taking advantage of the visible light-assisted method, our InAs NW photodetectors depict excellent performances, such as small dark current ($\sim \mu A$), low noise ($\sim 10^{-10}$ A/Hz¹⁄₂), high detectionivity ($\sim 10^{12}$ Jones for $\lambda = 2000$ nm), fast response ($\sim 80 \mu s$), and broad band detection. This method is suitable for all InAs NWs with rich surface states, regardless of the magnitude of the dark current (under zero gate voltage). It is noted that our experiments are conducted at 77 K. In fact, a higher temperature could also induce a similar phenomenon, for example, 160 K (see Supporting Information Figure S6). The difference is that a lower temperature would cause more electrons to be trapped and unable to be released, which leads to a higher Schottky barrier and a lower dark current. One question to which we need to pay more attention is that how long could the dark current stay at such a low level ($\sim \mu A$) after exposure to 450 nm light? We tested the device under chopped illumination of different power densities (see Supporting Information Figure S7) and found that the dark current changed little in more than 20 min. However, due to the feasibility of the reformation of high Schottky barriers by visible light, the time length of keeping at a low level seems to be not so important. To better understand the working mechanism, here we give a more detailed explanations on the formation and tunability of the Schottky contact. In our experiments, Cr/Au was used to make the electrodes. It is known that the work function of Cr is as low as 4.5 eV and the contact is usually Ohmic.¹¹,¹⁷ Though whether the high back-to-back Schottky barriers can be formed or not is mainly determined by the number of surface-trapped electrons, the contact metal is also a non-negligible factor. We also fabricated devices using Ni/Au as the electrodes. The similar MSM photodiodes’ formation with two high back-to-back barriers has also been found. However, it seems common that these MSM photodiodes have a higher breakdown voltage (see Supporting Information Figure S8). This is likely due to the high work function of Ni ($\sim 5.1$ eV), which may promote the formation of surface upward band bending.

In conclusion, we have demonstrated a visible light-assisted and single InAs NW-based MSM photodiode. The large back-to-back Schottky barriers induced by trapped electrons in PGL guarantee very low dark current and enhanced photoresponse to infrared light. A broad detection waveband from less than 1 $\mu m$ to more than 3 $\mu m$ has been achieved. Additionally, these photodiodes show high detectivity and fast response. Further experiments proved that our NPR is indeed from the native PGL itself and not the adsorption of gas molecules. A photocurrent mapping of the NW showed direct evidence that photocurrent mainly originates from the junction area. Our work successfully extended the detection waveband of single InAs NW photodetectors from the visible and near-infrared wavelengths to mid-infrared, and the devices show excellent performances for infrared detection. Through our studies, the great potential of InAs NWs for infrared optoelectronic applications was revealed.

**Methods. InAs NWs Synthesis.** The InAs NWs used in this work were synthesized on GaAs (111)B substrates in a Riber 32 MBE system through the vapor–liquid–solid (VLS) growth mechanism. The substrate surface was first deactivated at 570 °C for 15 min. Then, a GaAs buffer layer was grown for 15 min to ensure a smooth surface. Next, the preprocessed substrate was transferred to the MBE preparation chamber and an ultrathin Au film was then deposited on the substrate. Subsequently, to form gold droplets the Au-covered GaAs substrate was transferred back to the growth chamber and annealed for 5 min at 525 °C in an As₄ ambient atmosphere. Finally, the indium source was turned on to grow InAs NWs at 300 °C. Both the indium and As₄ fluxes were controlled by beam equivalent pressure (BEP). The indium/As₄ ratio was set at 1:20, and the growth time was 140 min. After the growth, the indium source and As₄ were subsequently switched off. Finally, the substrate was cooled to room temperature.

**Device Fabrication and Characterization.** InAs NWs were physically transferred to the SiO₂ (300 nm)/Si substrate. Then the source/drain (S/D) electrodes (15 nm Cr/45 nm Au) of each single InAs NW photodetector were fabricated using electron-beam lithography (EBL, JEOL 6510 with an NPGS System), metallization, and lift-off processes. Before metallization, the NWs were dipped into a 2% HF solution for approximately 10 s to remove the native oxide layer to ensure good contact between the metal electrode and the NW. The remaining oxide regions were protected by methyl methacrylate and poly(methyl methacrylate). All the devices were annealed at 200 °C in vacuum for 2 h before measurements. All the $I−V$ (current/voltage) characteristics were performed using a KEITHLEY 4200-SCS combined with a Lake Shore TTPX Probe Station. High time-resolution response curves were recorded by Tektronix MDO3014 mixed domain oscilloscope. HRTEM analysis was carried out on JEM2100F with acceleration voltage of 200 kV. The e-beam was spread carefully to avoid damage to InAs nanowires.

**ASSOCIATED CONTENT**

**Supporting Information**

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Additional figures (PDF)
Author Contributions

W.H. and X.C. conceived and supervised the research. H.F. fabricated the nanowire device. P.C. and X.Z. did the growth of the NWs. H.F. and P.W. performed the measurements. W.H. and H.F. wrote the paper. X.W. and C.L. conducted the HR-TEM and verified the crystal structure. H.T. performed the SEM characterization. All authors discussed the results and revised the manuscript.

Notes

The authors declare no competing financial interest.

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