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Near-infrared optical transitions in PdSe₂ phototransistors†

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We investigate electronic and optoelectronic properties of few-layer palladium diselenide (PdSe₂) phototransistors through spatially-resolved photocurrent measurements. A strong photocurrent resonance peak is observed at 1060 nm (1.17 eV), likely attributed to indirect optical transitions in few-layer PdSe₂. More interestingly, when the thickness of PdSe₂ flakes increases, more and more photocurrent resonance peaks appear in the near-infrared region, suggesting strong interlayer interactions in few-layer PdSe₂ help open up more optical transitions between the conduction and valence bands of PdSe₂. Moreover, gate-dependent measurements indicate that remarkable photocurrent responses at the junctions between PdSe₂ and metal electrodes primarily result from the photovoltaic effect when a PdSe₂ phototransistor is in the off-state and are partially attributed to the photothermoelectric effect when the device turns on. We also demonstrate PdSe₂ devices with a Seebeck coefficient as high as 74 μV K⁻¹ at room temperature, which is comparable with recent theoretical predications. Additionally, we find that the rise and decay time constants of PdSe₂ phototransistors are ~156 μs and ~163 μs, respectively, which are more than three orders of magnitude faster than previous PdSe₂ work and two orders of magnitude over other noble metal dichalcogenide phototransistors, offering new avenues for engineering future optoelectronics.

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Introduction

Despite the diverse array of electrical, mechanical, and optical properties in graphene, the lack of a sizeable band gap has limited its ability to be used in some electronic and optoelectronic applications.^{1,2} The unique structure as well as the extraordinary physical properties of graphene, owing largely to its two-dimensional (2D) nature, has generated intense interest to find other 2D materials with an appreciable band gap. One such class of materials is the transition metal dichalcogenide (TMD) family. These materials are characterized by the X–M–X structure, where the X represents a chalcogen atom (primarily

S, Se, Te), and the M is a transition metal from groups IV through VII or X. Though many TMDs are well understood in their bulk form, relatively few have received significant attention in their few-layer or monolayer form. The recent foray into the atomic thickness scale for these materials have revealed semiconductors (MoS₂, WSe₂, *etc.*), semimetals (WTe₂, *etc.*) and metals (VSe₂, *etc.*).^{3–5} The most notable and perhaps most studied 2D materials beyond graphene are group-VI TMDs such as MoS₂, MoSe₂, and WSe₂, which have shown fantastic electronic,^{6,7} optoelectronic,^{8,9} and valleytronic properties.^{10,11} The success of these materials has led to the exploration of other transition metals to form these compounds. Among group-VII TMDs, a high external quantum efficiency and a thickness-independent band gap of ReS₂ make it an attractive candidate for optoelectronics.¹² Additionally, a novel structure with mirror-symmetric single-crystal domains has been shown in monolayer ReSe₂ and is suggestive of novel anisotropic electronic and optoelectronic properties.¹³

More recently, the exploration of group-X TMDs (noble metal dichalcogenides) has begun. Excellent near-infrared (NIR) photodetection capabilities have been demonstrated in PtSe₂ field-effect transistors (FETs).¹⁴ And an ultrahigh photogain has been achieved in back-gate modulated PtS₂ devices.¹⁵ Of particular interest from this class of materials is PdSe₂. The

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high theoretically predicted mobility, an order of magnitude higher than Black Phosphorous (BP),¹⁶ air stability of the electronic properties, and thickness-dependent band gap ranging from ~ 0 eV in bulk to an indirect band gap of ~ 1.43 eV in a monolayer structure make it an interesting material to further probe its optoelectronic properties.^{17–19} Furthermore, the unique buckled pentagonal structure of PdSe₂ makes it a highly intriguing and desirable 2D material due to the resulting low-symmetry lattice structure. With the dominance of the hexagonal arrangement of atoms in popular 2D materials such as in MoS₂ and graphene or the buckled hexagonal arrangement of atoms in BP, the novel structure present in PdSe₂ offers many interesting opportunities.^{16–23}

In this study, electrical transport and optoelectronic measurements are performed on few-layer PdSe₂ phototransistors *via* scanning photocurrent microscopy. Our experimental results suggest that the strong photocurrent signals at metal–PdSe₂ junctions are mainly attributed to the photovoltaic effect (PVE) when PdSe₂ phototransistors turn off and partially related to the photothermoelectric effect (PTE) when the devices are in the on-state. We also demonstrate PdSe₂ devices with a Seebeck coefficient as high as $74 \mu\text{V K}^{-1}$ at room temperature, which is consistent with theoretical predications.^{18,20} More importantly, thickness-dependent photoresponse resonance peaks are observed in the NIR region, which likely results from indirect optical transitions in few-layer PdSe₂. Additionally, a fast response time is obtained ($\sim 156 \mu\text{s}$), which is an improvement of more than two orders of magnitude over previous reports for other TMD phototransistors and a three order of magnitude improvement over previous PdSe₂ work.^{12,13,15,23,24} This work offers new insight into the optoelectronic properties of PdSe₂ and opens up new avenues for

engineering future 2D noble metal dichalcogenide based electronics and optoelectronics.

Results and discussion

High-quality few-layer PdSe₂ flakes were mechanically exfoliated from a bulk crystal, and subsequently transferred to degenerately-doped Si substrates covered with a 270 nm SiO₂ layer. Thin PdSe₂ flakes (5–20 nm) were then identified by using optical microscopy and characterized with a Park-Systems XE-70 non-contact atomic force microscope (AFM). Finally, metal electrodes were fabricated onto the flakes by using standard electron-beam lithography and subsequent deposition of 10 nm Ti and 45 nm Au to create FETs where the degenerately-doped Si substrate was used as the back gate. Fig. 1a and b show a schematic and an optical micrograph, respectively, of an as-fabricated typical PdSe₂ transistor. The gold electrodes and few-layer PdSe₂ are outlined in gray and purple dashed lines, respectively. Electrical properties of the devices were measured in a Janis ST-500 Microscopy Cryostat under high vacuum ($\sim 10^{-6}$ Torr). Fig. 1c displays the gate-dependent electrical transport characteristics of a 9 nm (~ 15 layer) thick PdSe₂ transistor at room temperature. The device exhibits a predominately n-type behavior with an on/off ratio greater than $\sim 10^4$, which is better than or comparable to a previous report.²¹ The field-effect electron mobility of the device was calculated to be $\sim 92 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature by using the expression of $\mu_{\text{FET}} = L/W \times 1/C_{\text{bg}} \times dG/dV_{\text{G}}$, where L and W are the length and width of the channel, respectively; C_{bg} is the capacitance of the Si back gate; G is the conductance; and dG/dV_{G} is the slope of the G – V_{G} curve in the

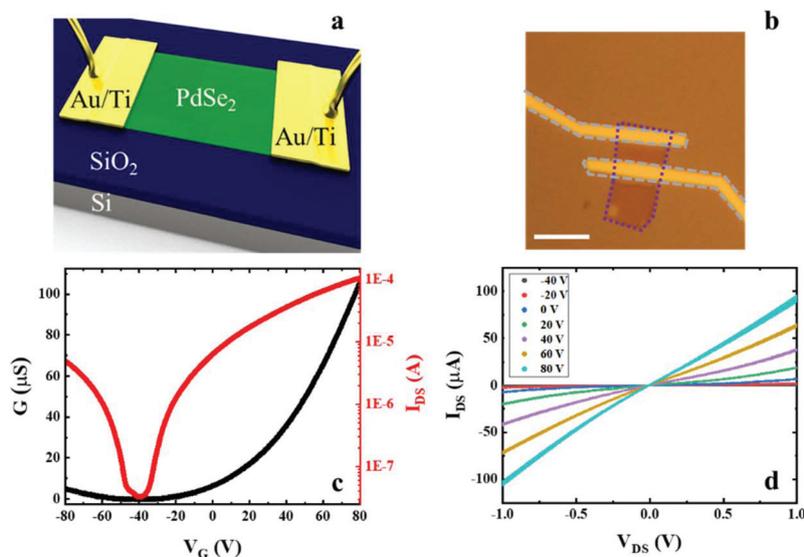


Fig. 1 (a) A schematic and (b) optical micrograph of an as-fabricated typical PdSe₂ phototransistor with gold electrodes and PdSe₂ channel outlined in gray and purple dashed lines, respectively. The scale bar in (b) is five microns. (c) Gate-dependent electrical transport of the PdSe₂ phototransistor. (d) The output characteristics of the device at various gate voltages.

linear region. Although the field-effect mobility observed in this device is significantly lower than theoretical predictions,^{16,17} it is comparable to or better than other experimental measurements.^{21,23} The observed field-effect mobility of the PdSe₂ device is likely limited by the presence of a substantial Schottky barrier (~ 180 meV) at the electrical contacts (see Fig. S1 of the ESI†). The linear output characteristic of the PdSe₂ device for gate voltages sweeping from -80 V to 80 V, as shown in Fig. 1d, can be attributed to thermally assisted tunneling through the Schottky barrier.²⁵

To investigate the photoresponse of the PdSe₂ device, spatially-resolved photocurrent measurements were performed in an Olympus microscopy setup. A continuous wave laser beam (NKT Photonics SuperK Supercontinuum Laser) was expanded and then focused by a $40\times$ Olympus objective (N.A. = 0.6) into a diffraction-limited spot (~ 1 μm) and scanned over the device using piezo-controlled mirrors with nanometer-scale spatial resolution. Fig. 2b shows the scanning photocurrent image of the PdSe₂ device under 1060 nm illumination at zero gate and zero drain-source bias, whose corresponding

reflection image was recorded simultaneously (Fig. 2a). The electrodes and PdSe₂ are outlined in gray and purple dashed lines, respectively. Strong photocurrent signals ($I_{\text{pc}} = I_{\text{laser}} - I_{\text{dark}}$) observed at the metal-PdSe₂ junctions are likely due to the creation of potential barriers from the Fermi level alignment at the junctions, which results in built-in electric fields that can efficiently separate photo-excited electron-hole pairs (EHPs) to generate photocurrent signals.²⁶

Furthermore, wavelength-dependent scanning photocurrent measurements were taken to investigate the photocurrent generation mechanisms in few-layer PdSe₂. Fig. 2c shows photocurrent responses of the PdSe₂ devices with different thicknesses. For thinner PdSe₂ phototransistors (e.g. 9 layers), a dominant resonance peak was observed at 1060 nm (1.17 eV), which is comparable to the indirect band gap of bi-layer PdSe₂ revealed by scanning tunneling spectroscopy (1.15 ± 0.07 eV).²⁷ Interestingly, when the thickness of PdSe₂ increases, more small peaks emerge around this near-IR region, such as peaks located at 1030 nm (1.20 eV) and 1090 nm (1.14 eV), respectively. These distinct peaks are likely

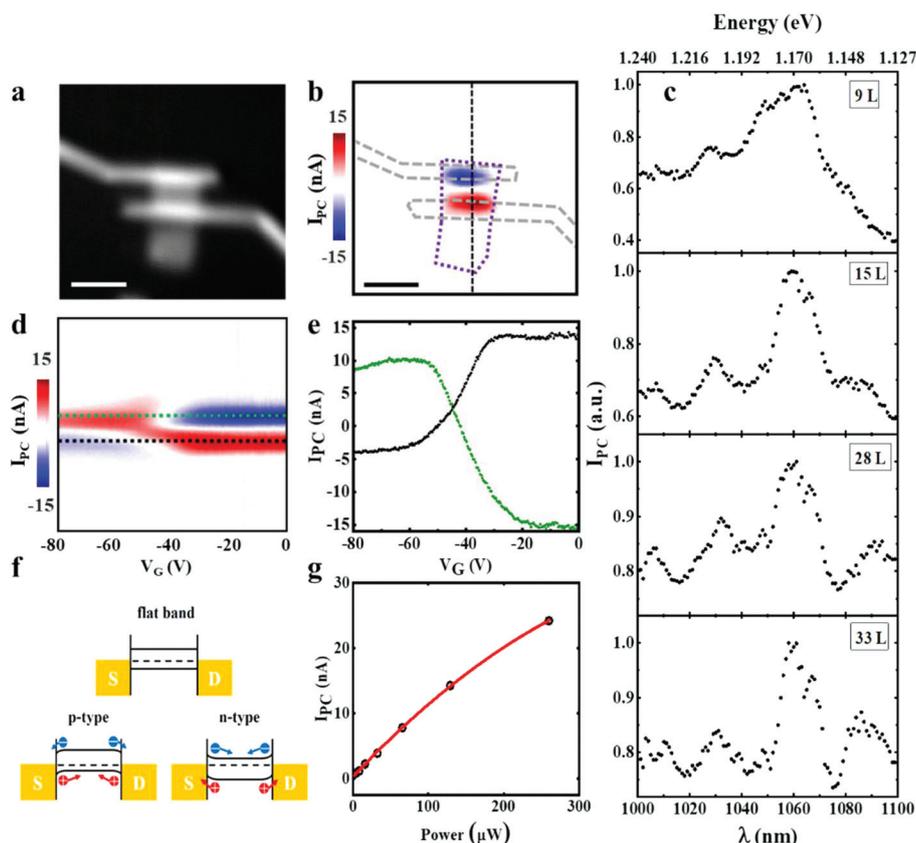


Fig. 2 (a) Reflection and (b) scanning photocurrent images of a typical PdSe₂ phototransistor under 1060 nm illumination. Metal electrodes and few-layer PdSe₂ channel are outlined by gray and purple dashed lines, respectively. The scale bars are five microns. (c) Normalized wavelength-dependent photocurrent responses of PdSe₂ phototransistors with different thickness. (d) Gate-dependent photocurrent signals along the black dashed cutline in (b) with a zero drain-source bias. (e) Green and black curves represent photocurrent responses along the green and black dashed lines in (d), respectively. (f) Schematic diagrams for photocurrent generation mechanisms for PdSe₂ phototransistors when the Fermi level moves from the valence band to the conduction band. (g) Power-dependent photocurrent behavior of the device, where the red solid line shows the trend line of $I_{\text{pc}} \propto P^\gamma$ with $\gamma = 0.85$.

attributed to different optical transitions between the local valence band maxima along the Γ -X line and the local conduction band minima located at the Γ -M line,¹⁹ which are affected by the thickness due to the strong interlayer coupling in the puckered pentagonal morphology of PdSe₂.²⁸ This may also explain the uncertainty of the few-layer PdSe₂ band gap characterization in previous optical absorption measurements (0.7–1.3 eV).¹⁷

To further study the photocurrent generation mechanisms, gate-dependent scanning photocurrent measurements were performed by sweeping the gate voltage from –80 V to 0 V while recording the photocurrent along the PdSe₂ channel (Fig. 2d). The photocurrent signal exhibits monotonic gate voltage dependence in the off-state (Fig. 2e), indicating that the PVE plays an important role in its photocurrent generation. Upon laser excitation, EHPs will be generated locally and driven in opposite directions by the built-in electric field owing to the Fermi level alignment that leads to Schottky barriers at metal–PdSe₂ junctions. Since the electronic energies are higher near the metal contacts than those in the middle of the PdSe₂ channel when the device is electrostatically n-doped (Fig. 2f, right), photo-excited electrons will be injected into the channel, leading to a negative/positive current flow in the drain/source electrode. The center of the channel shows negligible photocurrent response due to the relatively flat band structure at this location. When V_G decreases to –44 V, the flat band condition is reached (Fig. 2f, middle), resulting in negligible photocurrent response. Similarly, an opposite polarity of photocurrent response is observed when the device is operating in the p-type region (Fig. 2f, left).

Interestingly, the photocurrent signals start to saturate when V_G is larger than –20 V (Fig. 2e), which can be attributed to reduced gate-tunability of the band bending in the depletion regions as the Fermi level approaches the conduction band edge. In addition to photovoltaic mechanisms, other photocurrent generation mechanisms may also contribute to the photocurrent. Under illumination, the laser beam can also locally heat PdSe₂ to produce photocurrent response *via* either the photo-bolometric effect (PBE) or the PTE. When a laser beam scans over a PdSe₂ channel, the temperature of the channel increases due to light-induced heating, leading to the electrical conductance increase of the channel (Fig. S2a†).^{29,30} Therefore, PBE-induced photocurrent scales linearly with the drain–source bias. As shown in Fig. S2b,† the photocurrent response does not scale linearly with the drain–source bias, suggesting that the PTE may also contribute to the photocurrent generation. To further study the photocurrent generation mechanisms in metal–PdSe₂ junctions, we look into the spatially-resolved scanning photocurrent images of the PdSe₂ device. As shown in Fig. S2c,† a strong photocurrent “tail” is observed in the metal–PdSe₂ junction region, indicating that PTE also contributes to the photocurrent generation.^{31,32} Since the Seebeck coefficients of PdSe₂ (S_1) and the gold electrodes (S_2) are different, a photo-thermal voltage (V_{PTE}) across the metal–PdSe₂ junction exists:

$$V_{\text{PTE}} = (S_1 - S_2)\Delta T \quad (1)$$

From the Mott relation,^{32–34} we can obtain the Seebeck coefficient from

$$S = -\frac{\pi^2 k_B^2 T}{3e} \frac{1}{G} \frac{dG}{dV_G} \frac{dV_G}{dE} \Big|_{E=E_F} \quad (2)$$

where k_B is the Boltzmann constant, T is the temperature, e is the charge of an electron, and E_F is the Fermi energy. Here, the $\frac{dV_G}{dE} \Big|_{E=E_F}$ can be estimated as follows. When the Fermi level moves from the valence band to the conduction band, the barrier height E_b (the energy from the Fermi level to the valence band) changes linearly with V_G ($\delta E_b = e\alpha\delta V_G$), where α is a numerical constant that measures how effectively the gate voltage modulates the band energy.^{26,35} The measured shut-off gate voltages for p-type and n-type conductance are –60 V and –20 V, respectively (Fig. 1b). The band gap for few-layer PdSe₂ is 1.17 eV; therefore, the calculated $\alpha \approx 0.029$ can be used to infer the Seebeck coefficient. We obtained the maximum Seebeck coefficient of PdSe₂ ($\sim 74 \mu\text{V K}^{-1}$ at $V_G = -10$ V), which is consistent with the theoretically-predicted value of $\sim 200 \mu\text{V K}^{-1}$.²⁰ Therefore, photocurrent signals of PdSe₂ in the on-state may partially result from the PTE. Additionally, we performed power-dependent photocurrent measurements for the PdSe₂ device. As shown in Fig. 2g, the photocurrent signals have a nearly linear relationship with incident power ($I_{\text{PC}} \sim P^{0.85}$). A linear relationship is expected due to the increased generation of EHPs with increased number of incident photons. The slight deviation from the linear relationship may result from some defects and trap states in the PdSe₂ channel.^{8,13,36}

Next, we performed bias-dependent scanning photocurrent measurements on the PdSe₂ phototransistors at zero gate and different drain–source biases from –150 mV to 150 mV (Fig. 3a). Here the electrodes and few-layer PdSe₂ channel are outlined in gray and purple dashed lines, respectively. Regardless of the applied bias, strong photocurrent signals are observed at the metal–PdSe₂ junctions while the photocurrent response in the middle of the PdSe₂ channel is negligible. For comparison, corresponding horizontal cut lines along the channel (dashed lines in Fig. 3a) are shown in Fig. 3b. When a large positive bias is applied to the device, the positive photocurrent intensity at the source contact electrode increases. Similarly, the negative photocurrent signal in the drain contact electrode intensifies as the device is negatively biased. This increase in photoresponse under the application of a larger bias is primarily due to the enhancement of the electric field in the depletion region at the electrical contacts, which allows for more efficient separation and collection of the photo-excited EHPs. Since the slope of the electrostatic potential (or the local electric field) is roughly proportional to the photocurrent intensity, we numerically integrated the photocurrent curves in Fig. 3b to obtain the electrostatic potential (Fig. 3c). The potential change in different regions at various biases can be easily identified. Large potential drops near the electrode contact regions and relatively flat band in the middle of the PdSe₂ channel are observed, indicating the presence of contact

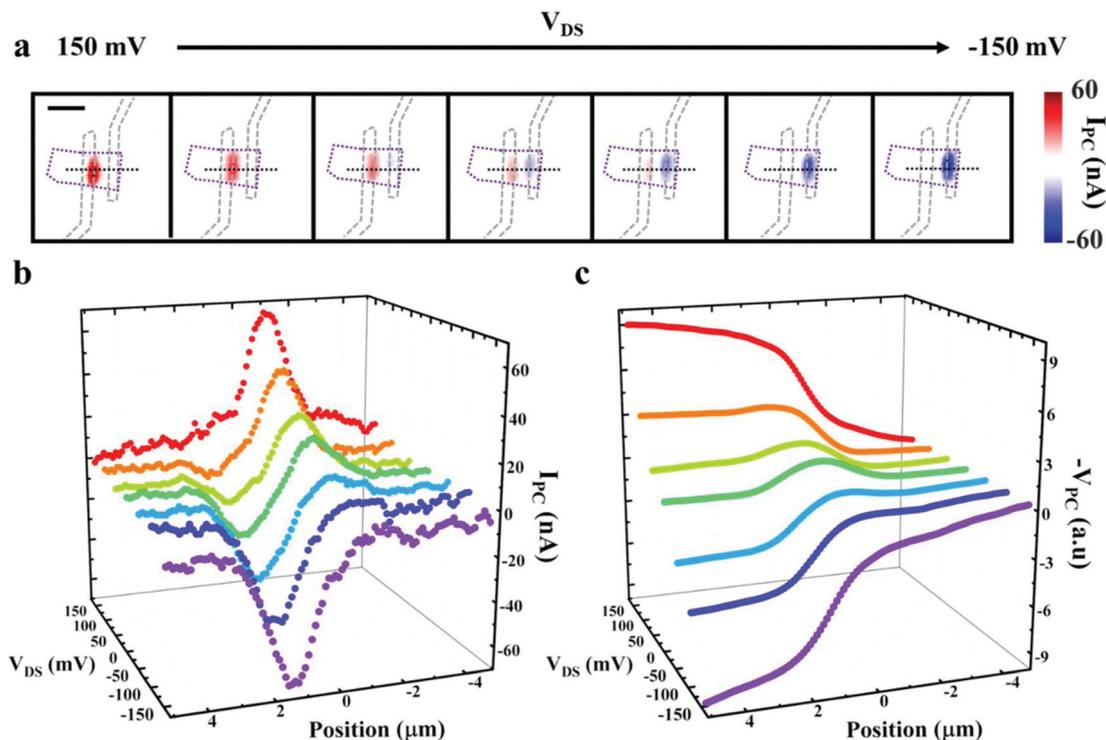


Fig. 3 (a) Scanning photocurrent images ($20\ \mu\text{m}$ by $20\ \mu\text{m}$) with drain–source biases sweeping from $-150\ \text{mV}$ to $150\ \text{mV}$. The scale bar is five microns. (b) Corresponding photocurrent intensity along the black dashed lines in (a). (c) Electrostatic potential computed by numerically integrating the photocurrent intensity curves from (b).

resistances due to the Schottky barriers between metal electrodes and the PdSe_2 channel. By selecting electrode metals with proper work functions and eliminating (or reducing) the Fermi level pinning, the Schottky barrier between electrode contacts and the PdSe_2 channel may be minimized.^{37–39}

We also study the photoresponse dynamics of PdSe_2 phototransistors through time-resolved scanning photocurrent measurements. In our experiment, an optical chopper was added to the system to apply ON/OFF light modulation while photocurrent signals were recorded as a function of time in

order to measure the rise and decay time constants, as shown in Fig. 4a. The rectangular excitation pulse generated by the chopper has a width of $\sim 2\ \text{ms}$ with a rise time of $\sim 100\ \text{ns}$. The device performed similarly over thousands of cycles of ON/OFF light modulation signifying the stability of the temporal response of the device. Fig. 4b shows three typical cycles. By applying a single exponential function to fit the rising and decaying regions of the curve, the obtained rise and decay time constants are $\sim 156\ \mu\text{s}$ and $\sim 163\ \mu\text{s}$ (Fig. 4c), respectively, which are more than three orders of magnitude faster than previous PdSe_2

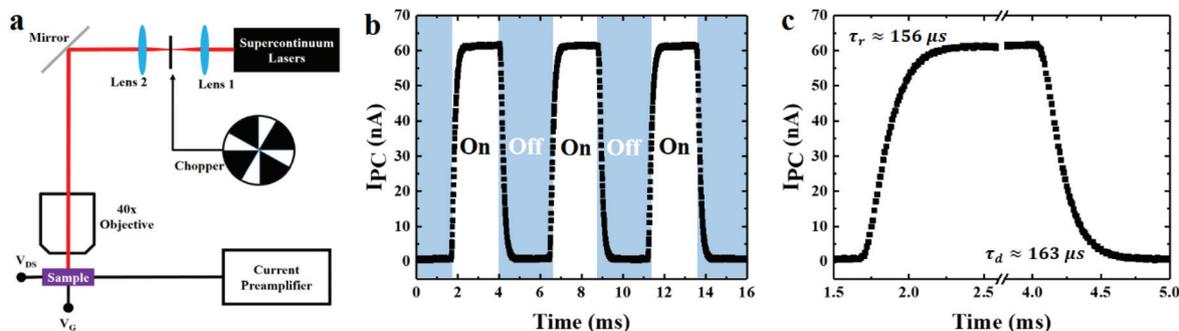


Fig. 4 (a) Schematic of the setup used to perform temporally-resolved photocurrent measurements. (b) Photocurrent signals as a function of time under $1060\ \text{nm}$ illumination. (c) Zoom-in of the rising and decaying regions of the photocurrent signals from (b) showing the rise and decay time constants of $\sim 156\ \mu\text{s}$ and $\sim 163\ \mu\text{s}$, respectively.

work and two orders of magnitude over other group-X TMD devices.^{12,13,15,23,24} These extremely fast response times are likely attributed to the high mobility of charge carriers in PdSe₂.^{16,40}

Conclusion

Through electrical transport and scanning photocurrent measurements, we demonstrate that strong photocurrent responses in PdSe₂ phototransistors primarily result from the PVE when the devices turn off and are partially related to the PTE when the devices are in the on-state. We also show PdSe₂ devices with Seebeck coefficients as high as 74 $\mu\text{V K}^{-1}$ at room temperature, which is consistent with theoretical predications. Moreover, thickness-dependent photocurrent resonance peaks are observed in the NIR region, which are likely attributed to the strong interlayer coupling-induced indirect optical transitions in few-layer PdSe₂. Additionally, a fast response time of $\sim 156 \mu\text{s}$ has been achieved in PdSe₂ phototransistors, which is more than three orders of magnitude faster than previous report and two orders of magnitude over other noble metal dichalcogenide devices. This work not only presents new insight into the optical transitions and photocurrent generation mechanisms of PdSe₂, but also opens up new avenues for engineering future noble metal dichalcogenide based optoelectronic devices.

Conflicts of interest

The authors declare no competing financial interests.

Acknowledgements

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