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# Long-Wave Infrared Photodetectors Based on 2D Platinum Diselenide atop Optical Cavity **Substrates**

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interaction in 2D materials and thus their photodetection performance in the LWIR spectral region. The PtSe<sub>2</sub> photoconductors fabricated on the TiO<sub>2</sub>/Au optical cavity substrate exhibit responsivities up to 54 mA/W to LWIR illumination at a wavelength of 8.35  $\mu$ m. Moreover, these devices show a fast photoresponse with a time constant of 54 ns to white light illumination. The findings of this study reveal the potential of multilayer PtSe<sub>2</sub> for fast and broadband photodetection from visible to LWIR wavelengths.

KEYWORDS: PtSe,, two-dimensional materials, noble-transition-metal dichalcogenides, light-matter interaction enhancement, light intensity and absorption, IR detectors, fast and broadband photodetection

nfrared (IR) photodetectors play a key role in a variety of areas including biomedical and thermal imaging, telecommunication, gas sensing, surveillance, search and rescue, environmental monitoring, and astronomy.<sup>1</sup> Mercury cadmium telluride (MCT), a composition-tunable direct bandgap semiconductor, has been extensively employed in mid-wave infrared (MWIR,  $3-8 \mu m$ ) and long-wave infrared (LWIR, 8–15  $\mu$ m) photodetectors.<sup>2,3</sup> Nevertheless, some issues such as difficulty of precise composition control and large-area nonuniformity result in a low-yield and complex fabrication process.<sup>2,4</sup> This has stimulated the search for alternative technologies for IR detection. Two-dimensional (2D) layered materials have been found to be promising for this purpose. $^{5-11}$ 

optical cavity substrate that enhances the light-matter

IR photodetectors based on graphene,<sup>12–17</sup> black phospho-rus (bP),<sup>18–21</sup> bPAs,<sup>22,23</sup> quasi-2D Te,<sup>24,25</sup> and SeTe<sup>26</sup> have been demonstrated. Recently, 2D noble-transition-metal dichalcogenides<sup>27</sup> including  $PtSe_{2}$ ,  $^{28-31}$  PdSe<sub>2</sub>,  $^{32-34}$  and PtS<sub>2</sub><sup>35</sup> have also been explored for IR detection. These materials possess appealing properties such as air stability, which is particularly an issue with bP;36 thickness-tunable bandgap; and relatively high carrier mobility.<sup>37</sup> Photodiodes

based on  $PtSe_2$  heterojunctions with Si,<sup>38-40</sup> Ge,<sup>41</sup> CdTe,<sup>42</sup> a perovskite,<sup>43</sup> PtS<sub>2</sub>,<sup>44</sup> and GaAs<sup>45</sup> have also been investigated for broadband photodetection up to short-wave infrared (SWIR, 1.4–3  $\mu$ m) wavelengths.

0

Time (ms)

Ab initio calculations reveal that PtSe<sub>2</sub> has a thicknessdependent band structure. Monolayer and bilayer PtSe2 are semiconductors with indirect bandgaps of ~1.2 and ~0.3 eV, respectively, and thicker flakes are semimetallic with zero bandgap.<sup>28,46</sup> Yu et al.<sup>28</sup> recently showed that the bandgap of bilayer PtSe2 can be further narrowed to the LWIR region (~0.11 eV) by introducing Se vacancies in the PtSe<sub>2</sub> crystals using a customized chemical vapor transport (CVT) system. Utilizing this material, they achieved a responsivity, detectivity, and rise/fall time of 4.5 A/W,  $7 \times 10^8$  Jones, and 1.2 ms,

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Figure 1. (a) Schematic of the infrared photodetector based on 2D materials atop the  $TiO_2/Au$  optical cavity substrate. (b) Normalized electric field intensity at the substrate surface in the absence of the 2D material for the  $SiO_2/Si$  ( $t_s = 300$  nm) substrate and  $TiO_2/Au$  substrate with different spacer thicknesses. (c and d) Normalized electric field intensity distribution in the cross section of the  $TiO_2/Au$  ( $t_s = 800$  nm) and  $SiO_2/Si$  ( $t_s = 300$  nm) substrates, respectively, in the absence of the 2D material at  $\lambda = 6.9 \,\mu$ m. The red dashed line represents the substrate surface, which lies at y = 0. (e) Same as b for the  $SiO_2/Si$  substrate with different  $SiO_2$  thicknesses.  $E_i$  is the norm of the incident electric field.

respectively, for illumination at a wavelength of 10  $\mu$ m. Despite the impressive device performance, from a manufacturing standpoint, it may prove challenging to produce PtSe<sub>2</sub> bilayers containing Se vacancies of the appropriate density in the reproducible manner required for devices such as image sensors.

In this study, we demonstrate, for the first time to the best of our knowledge, LWIR photodetectors based on multilayer  $PtSe_2$ . We employ a  $TiO_2/Au$  optical cavity substrate to boost the light-matter interaction in  $PtSe_2$  and enhance its LWIR detection performance. Optical cavity substrates have been previously used in graphene<sup>47</sup> devices in the MWIR,  $Te^{24}$  and  $SeTe^{26}$  devices in the SWIR, and  $MoS_2^{48}$  devices in the visible spectral regions. Our simulation and experimental results show that the performance of the 2D material-based LWIR detectors can be significantly improved using the optical cavity substrate. Device characterization reveals that the multilayer  $PtSe_2$  photodetectors exhibit a fast and broadband photoresponse.

# **RESULTS AND DISCUSSION**

The structure of the infrared photodetector based on 2D materials atop the  $TiO_2/Au$  optical cavity substrate is depicted in Figure 1a. The optical cavity substrate consists of a 120-nm-thick Au mirror, a  $TiO_2$  spacer layer with thickness  $t_{s}$ , and a 30-nm-thick  $Al_2O_3$  insulator film on a Si substrate. The reflected light from the Au mirror interferes constructively with the incident light at the substrate surface, leading to light intensity

enhancement. To maximize the light intensity at the substrate surface at wavelength  $\lambda$ , the spacer thickness  $(t_s)$  should be approximately equal to  $\lambda/(4n_s)$ , where  $n_s$  is the refractive index of the spacer. The Al<sub>2</sub>O<sub>3</sub> layer electrically insulates the 2D material sitting on the substrate from the spacer/mirror stack. This structure can be simultaneously used to apply a gate bias to the photodetector, where the Al<sub>2</sub>O<sub>3</sub> film acts as the gate dielectric and the Au/Si as the gate electrode. Here, we benchmark the TiO<sub>2</sub>/Au substrate against the commonly used SiO<sub>2</sub>/Si substrate, where the SiO<sub>2</sub> layer with thickness  $t_s$  acts as the insulator.

First, we conducted optical simulations to design the TiO<sub>2</sub>/Au optical cavity substrate and evaluate its performance. Figure 1b plots the electric field intensity (normalized to the incident intensity) at the substrate surface in the absence of the 2D material for the SiO<sub>2</sub>/Si ( $t_s = 300$  nm) substrate and TiO<sub>2</sub>/Au substrate with varying spacer thickness. As expected, the peak intensity wavelength red-shifts with an increase in the spacer thickness. The light intensity is considerably higher on the TiO<sub>2</sub>/Au substrate in comparison with the SiO<sub>2</sub>/Si substrate. For instance, at the peak intensity wavelength of 6.9  $\mu$ m for the TiO<sub>2</sub>/Au ( $t_s = 800$  nm) substrate, the light intensity is 10.4 times larger on the TiO<sub>2</sub>/Au substrate than that on the SiO<sub>2</sub>/Si substrate.

To further illustrate this, Figure 1c and d present the distribution of the electric field intensity in the cross section of the TiO<sub>2</sub>/Au ( $t_s = 800$  nm) and SiO<sub>2</sub>/Si ( $t_s = 300$  nm) substrates, respectively, in the absence of the 2D material at  $\lambda = 6.9 \ \mu$ m. The surface of the substrate at y = 0 is marked by a



Figure 2. (a and b) Optical power absorbed in the 2D material lying on the  $TiO_2/Au$  ( $t_s = 800 \text{ nm}$ ) and  $SiO_2/Si$  ( $t_s = 300 \text{ nm}$ ) substrates, respectively, with varying thickness ( $t_{2D}$ ) and refractive index (n and k) of the 2D material. (c-e) Optical power absorbed and absorption enhancement in the 2D material lying on the  $TiO_2/Au$  ( $t_s = 800 \text{ nm}$ ) and  $SiO_2/Si$  ( $t_s = 300 \text{ nm}$ ) substrates as functions of n and k with  $t_{2D} = 10$ , 20, and 100 nm, respectively, at  $\lambda = 8.35 \,\mu$ m. The absorption enhancement is defined as the ratio of the 2D material absorption with the  $TiO_2/Au$  substrate to that with the  $SiO_2/Si$  substrate.

red dashed line. These profiles indicate that for the TiO<sub>2</sub>/Au substrate with proper spacer thickness, the maximum intensity  $(|E|^2/|E_i|^2 = 3.76)$  lies at the substrate surface, while for the SiO<sub>2</sub>/Si substrate, this maximum  $(|E|^2/|E_i|^2 = 2.37)$  occurs at  $y = 1.43 \ \mu m$  (*i.e.*, at 1.43  $\ \mu m$  above the substrate surface). It should also be noted that the light intensity at the surface of the SiO<sub>2</sub>/Si substrate is smaller than the incident intensity ( $|E|^2/|E_i|^2 = 0.36$ ). This comes from the destructive interference between the incident and reflected waves at the substrate surface.

It is noteworthy that the electric field intensity at the surface of the SiO<sub>2</sub>/Si substrate can also be optimized in the MWIR region by adjusting the SiO<sub>2</sub> thickness, as shown in Figure 1e. However, as observed in this figure, SiO<sub>2</sub>/Si is not an appropriate substrate for the LWIR region, since the high absorption of SiO<sub>2</sub> at these wavelengths leads to low light intensity at the substrate surface. Furthermore, other studies typically use an SiO<sub>2</sub>/Si substrate with a SiO<sub>2</sub> thickness of 300  $m^{17,23,29,30,32,44}$  or less<sup>12,14–16,20,22,28,33,34</sup> in the 2D materialbased photodetectors, regardless of the operating wavelength. Accordingly, we consider an SiO<sub>2</sub> thickness of 300 nm in the following simulations and experiments.

The time-averaged optical power density absorbed in the 2D material sitting on the substrate can be expressed as  $^{49}$ 

$$\left\langle \overrightarrow{E(t)} \cdot \frac{\partial \overrightarrow{P(t)}}{\partial t} \right\rangle = \frac{1}{2} \omega \epsilon'' |\vec{E}|^2 = \epsilon_0 \omega n k |\vec{E}|^2$$

where  $\vec{E}$  denotes the electric field,  $\vec{P}$  electric polarization,  $\omega$  angular frequency,  $\epsilon = \epsilon' + i\epsilon'' = \epsilon_0(n + ik)^2$  2D material's

permittivity, and  $\epsilon_0$  vacuum permittivity. *n* and *k* are the real and imaginary parts of the 2D material refractive index, respectively. According to this equation, the optical power absorbed in the 2D material is proportional to the electric field intensity, *n*, and *k*.

The above discussions imply that a thin 2D material with negligible influence on the light intensity distribution, such as monolayer graphene, would absorb ~10.4 times more power on the TiO<sub>2</sub>/Au ( $t_s = 800$  nm) substrate compared with the SiO<sub>2</sub>/Si ( $t_s = 300$  nm) substrate at  $\lambda = 6.9 \ \mu$ m. However, this is not true for a thick multilayer 2D material with significant reflection and absorption. Next, we investigate the effect of the thickness ( $t_{2D}$ ) and complex refractive index (n + ik) of the 2D material on the light absorption in the 2D material sitting on the TiO<sub>2</sub>/Au and SiO<sub>2</sub>/Si substrates.

Figure 2a presents the optical power (normalized to the source power) absorbed in the 2D material sitting on the  $TiO_2/Au$  ( $t_s = 800 \text{ nm}$ ) substrate as a function of wavelength for different  $t_{2D}$ , n, and k values. The absorbed power was obtained by calculating the net power flow into a surface enclosing the 2D material. It is observed that the 2D material absorption increases with an increase in its thickness. There is also a red-shift in the peak absorption wavelength with the increase in thickness. At constant thickness, larger n and larger k values lead to larger absorption. Also, there is a red-shift and a blue-shift in the peak absorption wavelength with the increase in n and k, respectively. The optical power absorbed in the 2D material on the SiO<sub>2</sub>/Si ( $t_s = 300 \text{ nm}$ ) substrate is plotted in Figure 2b for the same  $t_{2D}$ , n, and k values as Figure 2a. Here again, the power absorbed in the 2D material



Figure 3. (a) Top and (b) side views of the multilayer 1T-PtSe<sub>2</sub> crystal structure with AA stacking order. (c) Optical micrograph of a PtSe<sub>2</sub> photodetector fabricated on the TiO<sub>2</sub>/Au ( $t_s = 800$  nm) optical cavity substrate. (d) Atomic force micrograph of the PtSe<sub>2</sub> flake in panel c. (e) Drain current *vs* gate voltage characteristic of the PtSe<sub>2</sub> device.



Figure 4. Photoresponse of a PtSe<sub>2</sub> photodetector on the TiO<sub>2</sub>/Au ( $t_s = 800 \text{ nm}$ ) substrate to the 8.35  $\mu$ m laser. (a) Photoresponse with the laser modulated at 700 Hz at the incident power density of 43.5 W/cm<sup>2</sup>. (b) Photocurrent as a function of laser power density. (c) Responsivity and detectivity as functions of laser power density. The drain bias was  $V_d = 50 \text{ mV}$ , and the gate electrode was disconnected in the above measurements.

increases with an increase in its thickness and refractive index values. Also, we note that the 2D material absorption is greater on the  $TiO_2/Au$  than that on the  $SiO_2/Si$  substrate over a wide wavelength range.

To further study the 2D material absorption enhancement provided by the TiO<sub>2</sub>/Au substrate, Figure 2c-e illustrate the power absorbed and absorption enhancement in the 2D material at  $\lambda = 8.35 \ \mu m$  for a wide range of *n* and *k* values on the TiO<sub>2</sub>/Au ( $t_s = 800 \ nm$ ) and SiO<sub>2</sub>/Si ( $t_s = 300 \ nm$ ) substrates for  $t_{2D} = 10, 20$ , and 100 nm, respectively. The same results for  $t_{2D} = 50 \ nm$  are shown in the Supporting Information, Figure S1. Here, the absorption enhancement is defined as the ratio of the 2D material absorption with the  $\text{TiO}_2/\text{Au}$  substrate to that with the  $\text{SiO}_2/\text{Si}$  substrate. The absorption increases almost monotonically with an increase in n and k for  $t_{2D} = 10$  and 20 nm for both substrates. However, this is not the case for the thicker 2D materials ( $t_{2D} = 50$  and 100 nm); for instance, the 100-nm-thick 2D material on the  $\text{TiO}_2/\text{Au}$  substrate has a maximum absorption of 94.8% at (n, k) = (3.75, 1.75). Regardless of  $t_{2D}$ , the absorption enhancement is maximum for low-absorbance materials (small k values), and it drops with an increase in k. Also, the absorption enhancement is generally higher for thinner 2D materials. Here we observe a maximum absorption enhancement of 9.6 for (n,



Figure 5. (a) Drain current as a function of temperature for a PtSe<sub>2</sub> device on the SiO<sub>2</sub>/Si ( $t_s = 300$  nm) substrate in the dark. (b) Responsivity and detectivity of a PtSe<sub>2</sub> photodetector on the TiO<sub>2</sub>/Au ( $t_s = 800$  nm) substrate to the 8.35  $\mu$ m laser as functions of laser power density at room and cryogenic temperatures. The drain bias was  $V_d = 50$  mV, and the gate electrode was disconnected in the above measurements.



Figure 6. (a) Responsivity and (b) detectivity of different PtSe<sub>2</sub> devices to the 8.35  $\mu$ m light on the TiO<sub>2</sub>/Au ( $t_s = 800 \text{ nm}$ ) and SiO<sub>2</sub>/Si ( $t_s = 300 \text{ nm}$ ) substrates. Each bar represents a device. The drain bias was  $V_d = 50 \text{ mV}$ , and the gate electrode was disconnected in the above measurements.

k) = (6, 0.1) for  $t_{2D}$  = 20 nm and a minimum absorption enhancement of 1.84 for (n, k) = (6, 5) for  $t_{2D}$  = 100 nm.

Next, we fabricated and characterized PtSe<sub>2</sub> photoconductors on TiO<sub>2</sub>/Au ( $t_s = 800$  nm) and SiO<sub>2</sub>/Si ( $t_s = 300$  nm) substrates. The stable octahedral 1T crystal structure of PtSe<sub>2</sub> is depicted in Figure 3a. PtSe<sub>2</sub> layers are stacked in the AA arrangement, as shown in Figure 3b, to form multilayer PtSe2.29 Here, PtSe2 photodetectors were prepared by the mechanical exfoliation of the PtSe<sub>2</sub> flakes onto the substrate, followed by the patterning and deposition of the Cr/Au source and drain electrodes. Multilayer PtSe<sub>2</sub> flakes with thicknesses in the range of  $\sim 10-70$  nm were chosen for the fabrication of these photodetectors. PtSe<sub>2</sub> flakes thinner than ~10 nm and large enough for device fabrication ( $\geq 3 \mu m$ ) were very scarce. Figure 3c presents an optical microscope image of a PtSe<sub>2</sub> photodetector on the TiO2/Au substrate. An atomic force microscope (AFM) image of this PtSe<sub>2</sub> flake is included in Figure 3d, which shows that this flake has an uneven thickness ranging from 56 to 71 nm. The drain current-gate voltage  $(I_d - V_g)$  characteristic of this device is shown in Figure 3e. The drain-source voltage  $(V_d)$  was kept at 50 mV. Multilayer PtSe<sub>2</sub> is a semimetallic material; hence, there is no obvious gate control over drain current. Nonetheless, we observed weak ptype and ambipolar behavior in few-layer PtSe<sub>2</sub> flakes with thicknesses below 5 nm. Figure S2 in the Supporting Information displays examples of  $I_d - V_g$  characteristics of few-layer PtSe<sub>2</sub> flakes on the SiO<sub>2</sub>/Si substrate.

The photoresponse of the PtSe<sub>2</sub> device on the TiO<sub>2</sub>/Au ( $t_s =$ 800 nm) substrate to the LWIR laser ( $\lambda = 8.35 \ \mu m$ ) is shown in Figure 4a, where the laser was pulsed at 700 Hz and the incident power density was 43.5 W/cm<sup>2</sup>. The drain bias was  $V_{\rm d}$ = 50 mV, and the gate electrode was disconnected. All measurements were taken at room temperature unless stated otherwise. A photocurrent  $(I_{ph} = I_{light} - I_{dark})$  of 184 nA is observed. Figure 4b plots the photocurrent as a function of laser power density for this device. We observe a linear behavior, which is desirable for photodetectors. The responsivity of a photodetector is defined as  $R = I_{\rm ph}/P_{\rm in}$ , where  $P_{in}$  is the power incident on the device. Figure 4c shows the responsivity of this device as a function of laser power density. A maximum responsivity of 54.3 mA/W is measured. The detectivity, which represents the photodetector's ability to differentiate the signal from noise, is defined as

$$D^* = \frac{\sqrt{A\Delta f}}{\text{NEP}} = \frac{R\sqrt{A\Delta f}}{\sqrt{I_n^2}}$$

where A is the device area,  $\Delta f$  sampling bandwidth, NEP noise equivalent power, R responsivity, and  $I_n$  noise current. Assuming that the total noise of the photodetector is dominated by the shot noise from the dark current, the noise current can be approximated as  $\overline{I_n}^2 \approx 2eI_{\text{dark}}\Delta f$ .<sup>5,24</sup> This leads us to



Figure 7. (a) Photoresponse of a PtSe<sub>2</sub> photodetector on the SiO<sub>2</sub>/Si ( $t_s = 300 \text{ nm}$ ) substrate to 532 and 1550 nm lasers at incident power densities of 5661 and 5242 W/cm<sup>2</sup>, respectively. The laser beams were chopped at 700 Hz. (b) Photoresponse of a PtSe<sub>2</sub> photodetector on the SiO<sub>2</sub>/Si ( $t_s = 300 \text{ nm}$ ) substrate to the pulsed white light ( $\lambda = 450-2400 \text{ nm}$ ) laser with a pulse width of 2 ns, along with the analog pulse signal of the laser. The drain bias was  $V_d = 50 \text{ mV}$ , and the gate electrode was disconnected in the above measurements.

$$D^* \approx \frac{R\sqrt{A}}{\sqrt{2eI_{\text{dark}}}}$$

where *e* is the unit charge and  $I_{dark}$  is the dark current. The detectivity of this device, calculated using the latter equation, is reported in Figure 4c. It shows a maximum detectivity of 2.5 ×  $10^6$  Jones (cm $\sqrt{Hz}/W$ ).

To understand the photodetection mechanism in these devices, we studied the temperature dependence of the drain current in the dark. Figure 5a plots the dark current as a function of temperature for a PtSe<sub>2</sub> device on the SiO<sub>2</sub>/Si ( $t_s$  = 300 nm) substrate. The same type of measurement for another device is shown in the Supporting Information, Figure S3. As expected for a semimetallic material, the current decreases with an increase in temperature. This is consistent with the findings of ref 29. Hence, the photoresponse does not originate from the temperature rise induced by the illumination (bolometric effect) and is instead attributed to the generation of photocarriers and their collection by the electrodes (photoconductive effect). The effect of cryogenic cooling on device performance was also investigated. Figure 5b plots the responsivity and detectivity of a PtSe<sub>2</sub> photodetector on the  $TiO_2/Au$  ( $t_s = 800$  nm) substrate to the 8.35  $\mu$ m laser at different temperatures. Both the light and dark currents increase as the device is cooled from 295 to 100 K. The dark current increases from 162 to 294  $\mu$ A. The photocurrent and responsivity are enhanced by 3.15 times and detectivity by 2.34 times.

Several PtSe<sub>2</sub> photodetectors were fabricated on the TiO<sub>2</sub>/ Au ( $t_s = 800$  nm) and SiO<sub>2</sub>/Si ( $t_s = 300$  nm) substrates. The responsivity and detectivity of these devices to the 8.35  $\mu$ m laser are compared in Figure 6a and b, respectively. Each bar represents a device. As expected, the photodetectors on the TiO<sub>2</sub>/Au substrate generally perform better than those on the SiO<sub>2</sub>/Si substrate. The responsivity and detectivity of the best device on the TiO<sub>2</sub>/Au substrate are respectively 2.7 and 2.6 times higher than those of the best device on the SiO<sub>2</sub>/Si substrate. Also, the average values of responsivity and detectivity for the devices on the TiO<sub>2</sub>/Au substrate are respectively 4.5 and 3 times higher than those for the devices on  $SiO_2/Si$ .

As shown in Figure 6a, the best device on the SiO<sub>2</sub>/Si substrate shows a responsivity of 20 mA/W to the LWIR light ( $\lambda = 8.35 \ \mu$ m). We also measured the response of this device to visible ( $\lambda = 532 \ \text{nm}$ ) and SWIR ( $\lambda = 1550 \ \text{nm}$ ) light; the results are plotted in Figure 7a. The visible and SWIR laser beams were chopped at 700 Hz. This device shows responsivity values of 2.1 and 1.1 mA/W to the 532 and 1550 nm light, respectively.

The rise/fall times of the photoresponse plotted in Figure 4a and Figure 7a are limited by the measurement setup, *i.e.*, the speed of the laser in Figure 4a and the chopping process in Figure 7a, rather than the photodetector speed. To characterize the response time of the PtSe<sub>2</sub> photodetectors, a device on the SiO<sub>2</sub>/Si substrate was illuminated by a pulsed white light source ( $\lambda = 450-2400$  nm) with a pulse width of ~2 ns. Figure 7b presents the photoresponse of the device, together with the analog pulse signal of the laser. This device responds to the light pulse with a time constant of 54 ns. From another viewpoint, the impulse response of the photodetector decays exponentially, *i.e.*,  $e^{-t/\tau}$ , where  $\tau \approx 54$  ns is the recombination lifetime of the photogenerated carriers.<sup>50</sup>

#### **CONCLUSION**

In summary, we demonstrated fast and broadband (visible to LWIR) photodetectors based on multilayer PtSe<sub>2</sub>, which was mechanically exfoliated from standard CVT-grown crystals. Moreover, we designed and fabricated an optical cavity substrate to boost the light–matter interaction in 2D materials in the LWIR spectral region. We employed this TiO<sub>2</sub>/Au optical cavity substrate to enhance the performance of PtSe<sub>2</sub> photodetectors. These devices show a maximum responsivity of 54 mA/W and detectivity of 2.5 × 10<sup>6</sup> Jones to LWIR illumination (at a wavelength of 8.35  $\mu$ m) at room temperature. Higher responsivity and detectivity values were reported in ref 28 for bilayer PtSe<sub>2</sub> containing Se vacancies. Our devices, however, are much faster and more practical from a manufacturing perspective. Two-dimensional materials have shown great potential for IR photodetection lately, and this

study highlights the significant effect of the substrate on the performance of these devices.

#### **METHODS**

**Simulation.** Maxwell's equations were solved employing the finitedifference time-domain (FDTD) technique,<sup>51</sup> implemented in the Lumerical FDTD Solutions software package. The light source was a broadband ( $\lambda = 5-13 \mu$ m) linearly polarized plane wave at normal incidence. Perfectly matched layers (PMLs) were utilized to describe the boundary conditions in the light propagation direction. Periodic boundary conditions were implemented in the direction perpendicular to light propagation. The mesh size was 1 nm around the finest features. The wavelength-dependent complex refractive index values of TiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and SiO<sub>2</sub> were taken from ref 52, and those of Au and Si were taken from ref 53.

**Fabrication.** The fabrication of the  $TiO_2/Au$  optical cavity substrate started with e-beam evaporation of a 20/120 nm Cr/Au reflector on a Si substrate. Next, a 1 nm Ti layer was e-beam evaporated to act as the seed layer for the  $TiO_2$  deposition. This was followed by the deposition of the 800 nm  $TiO_2$  spacer *via* reactive sputtering physical vapor deposition. Finally, a 30 nm  $Al_2O_3$  insulator film was deposited using atomic layer deposition (ALD). For the  $SiO_2/Si$  substrate, the thermally grown  $SiO_2$  insulator layer was 300 nm thick.

CVT-grown PtSe<sub>2</sub> crystals were purchased from HQ Graphene. PtSe<sub>2</sub> flakes were deposited onto the substrate by exfoliating the crystals using a sticky tape. Next, the source/drain electrodes (Cr/Au 20/80 nm) were fabricated using the photolithography/e-beam evaporation/lift-off method. Subsequently, the thickness of the PtSe<sub>2</sub> flakes was measured using an atomic force microscope (Cypher AFM, Asylum Research) in tapping mode. Finally, the chips were wire-bonded into chip carriers for electrical and optical characterization.

Characterization. The electrical characteristics were measured using a pair of source measure units (2450 SourceMeter, Keithley Instruments) in a two-probe configuration. In the photoresponse experiments, the 8.35  $\mu$ m quantum cascade laser (QCL) was directly modulated at 700 Hz. The beams from 532 and 1550 nm laser diodes were mechanically chopped at 700 Hz. A thermal power sensor (S401C, Thorlabs) was used to measure the optical power of the lasers. The spot sizes of the lasers were measured using the knife-edge method and the power meter, and thus the power densities were obtained. The signal from the photodetector was amplified using a low-noise current preamplifier (SR570, Stanford Research Systems), and the output was monitored on an oscilloscope. The drain bias was applied using the current preamplifier. Temperature-dependent measurements were taken in a customized cryostat (Janis Research) with a ZnSe window. The response time was characterized by illuminating the device with 2-ns-wide pulses from a supercontinuum white light laser (SuperK Compact, NKT Photonics). A high-speed current amplifier (DHPCA-100, FEMTO) with a bandwidth of ~170 MHz was used in this case.

# ASSOCIATED CONTENT

#### **Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsnano.0c09739.

Optical power absorbed in the 50-nm-thick 2D material,  $I_d-V_g$  characteristics of few-layer PtSe<sub>2</sub> devices, temperature dependence of dark current for a PtSe<sub>2</sub> device (PDF)

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# Notes

The authors declare no competing financial interest.

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