High-sensitivity broad-spectrum photodetectors with detection capabilities ranging from ultraviolet to infrared have attracted significant attention for their application as photodetectors. They can be used as a receiver for all applications in optical communication for covering a wide spectral range with a single photodetector, which can significantly lower the overall system cost. In this study, by constructing an avalanche photodetector (APD) fabricated with 2D WSe$_2$, a high photoresponsivity of over $10^7$ A W$^{-1}$ for a broad spectrum of 405–1310 nm is achieved under an electric field higher than the critical field for the avalanche multiplication of 35 kV cm$^{-1}$, overcoming the limitation of the detectable wavelength induced by the energy bandgap of the material. The benchmark in terms of the photocurrent-to-dark current ratio and responsivity over a wide wavelength range demonstrates that the fabricated WSe$_2$ APD outperforms the reported 2D layered material-based APDs and reported WSe$_2$ photodetectors. The obtained results can be attributed to the high-gain mechanism via avalanche multiplication and phonon-assisted photogeneration in WSe$_2$, enabling efficient photodetection beyond sub-bandgap wavelengths. This result provides a promising general approach for developing a single photodetector that can cover a broad spectral range with a high sensitivity for future optical communication.

1. Introduction

Photodetectors that convert optical signals into electrical signals have various uses. Beyond medical X-rays and visible light sensors, it is emerging as an important component in new applications, such as medical near-infrared imaging and far-infrared surveillance.[1–3] Until now, separate photodetectors covering different spectral ranges have been used for each purpose, but recently, the need for a broad-spectrum photodetector has increased for long-haul applications which use a wide spectral wavelength of light. Next-generation communication receivers require a single photodetector that can detect multiple spectral bands ranging from ultraviolet (UV) to infrared (IR) light.[4] Using a single photodetector with broad-spectrum sensing can make the device unusually small, light, and low powered and help reduce inventories and decrease overall system costs.

Historically, photodetection has relied on the bandgap of semiconductors to convert photons with different energies into electrical signals. Light with a wavelength of $\lambda = \frac{hc}{E_g}$ is required to raise electrons from the valence band to the conduction band, where $h$ is the Planck constant, $c$ is the speed of light, and $E_g$ is the bandgap of the material. Detection of ultraviolet (UV) and visible light has been enabled by conventional materials such as Si and InGaAs, which have high performance and low-cost fabrication.[5,6] However, these materials fail to absorb light in the infrared spectrum with a wavelength of over 1.1 $\mu$m and have a limitation of low absorption for the entire spectrum.[7] Therefore, several efforts have been made to discover new materials and structures that expand the absorption spectrum or improve device sensitivity in the entire spectral range by self-signal amplification via carrier multiplication.

2D layered materials, with advantages of thickness-dependent tunable bandgap, easy processing, and strong light interaction, have recently attracted enormous research interests as promising materials for photodetector.[8] The most widely studied 2D layered materials, n-MoS$_2$ and p-WSe$_2$, cover light up to 700 and 980 nm, respectively,[9,10] this can be inferred from the relationship between the bandgap of the material and detectable wavelength of light. Their spectral range corresponds to the visible light range; thus, their application in optoelectronics has attracted much attention. However, for use as a photodetector, it has a low optical absorption of $\approx 10%$[11] because of its atomic thickness and limited spectral range coverage. As breakthroughs, new device architectures such as complicated
heterostructures[12] or multiperiod grating structures[13] have been proposed. High-sensitivity and broad-spectrum photodetection was achieved by Jeon et al.[13] who demonstrated light-to-dark ratio of $>10^8$ only to a broad spectral range (405–1310 nm) by transition-metal-carbide multiperiod gratings. However, these complex structures overshadow the ease of processing 2D layered materials.

Recently, avalanche photodetectors (APDs) with high sensitivity have been reported, overcoming the intrinsic limitations of 2D layered materials. Two distinct approaches can be identified from previous works: use of a simple field-effect transistor (FET) structure based on the impact ionization properties of 2D materials[14, 15, 16–17] and the design of complex device structures that enhance avalanche multiplication effects through the use of a plasmonic grating[18] or heterojunction structures.[19, 20]

A comparison of the photodetection performances of these devices with ours is presented in Table S1 (Supporting Information). It is known that 2D layered materials are promising for impact ionization underpinning avalanche multiplication owing to the strong Coulomb interaction-induced quantum confinement effect and the modulation of their band structures. Avalanche photodetection via impact ionization plays a substantial role in amplifying signals by the device. Carriers, including photoinduced carriers, accelerate sufficiently under a high electric field above the critical electric field ($E_{CR}$). Carriers with a high kinetic energy collide with the lattice to generate free carriers. These newly generated carriers undergo the same process, causing an increase in the number of carriers, which is similar to an avalanche. Based on this mechanism, the APD acts as a photomultiplier with carrier multiplication and exhibits high photocurrent and photoresponsivity. A MoS$_2$ APD[14] demonstrated high responsivity ($>10^7$ A W$^{-1}$) and detectivity ($>10^{16}$ Jones) at a high electric field of over 350 kV cm$^{-1}$ ($E_{CR}$ of MoS$_2$), and a BP-Au NPs APD[15] exhibited a responsivity of 160 A W$^{-1}$ with a low $E_{CR}$ of 5 kV cm$^{-1}$. However, from an electronic point of view, a high bias is required for the operation of APDs so that their $E_{CR}$ is exceeded. Therefore, efforts to reduce this bias are essential for low-power devices, and it is important to select a material with a low $E_{CR}$.

In this study, we demonstrated a high-performance APD by simply forming metal contacts on a mechanically exfoliated multilayer WSe$_2$ on a SiO$_2$/Si substrate. WSe$_2$ exhibits superior avalanche multiplication characteristics, its impact ionization is triggered at extremely low electric fields (35 kV cm$^{-1}$), and it demonstrates a record-high multiplication factor of $>10^5$. Owing to its great carrier multiplication, the photogenerated carriers are also multiplied by a high-gain mechanism in the avalanche regime (i.e., $E > E_{CR}$), allowing the device to have a record-high photoresponsivity of $\sim 10^8$ A W$^{-1}$. Furthermore, we investigated its performance at various wavelengths of light, ranging from 405 to 1310 nm. In the low-bias regime of $E < E_{CR}$, it responded with a responsivity of $\sim 10^3$ A W$^{-1}$ only to light with a short wavelength of 1240 nm or less, similar to previous reports.[21–27] On the other hand, in the high-bias regime (i.e., avalanche regime) of $E > E_{CR}$, the responsivities for all investigated wavelengths were enhanced up to $10^7$ W A$^{-1}$, indicating that it was possible to detect broad-spectrum light with a single photodetector. This nearly bandgap-independent broadband photodetection is achieved by amplifying the number of phonon-assisted photogenerated carriers through avalanche multiplication.

In general, the direct optical transition less than the bandgap energy is not allowed in semiconductors and insulators. However, the indirect optical transition with less than the direct interband transition energy is possible when phonons are emitted or absorbed, and this phonon-assisted optical transition is responsible for most of the absorption near the threshold. In this indirect transition, the creation of electron–hole pairs does not violate the energy and momentum conservation when phonons of appropriate wave vectors are created or destroyed. In our device, a band bending with an applied bias voltage leads to phonon-assisted indirect optical transitions. Upon the illumination of light having smaller energy than the bandgap, the phonon-assisted light absorption is allowed to produce electron–hole pairs. More importantly, the absorption of longer wavelength light is possible with increasing bias, indicating strongly enhanced absorption processes with the assistance of phonon modes under large electric fields. Thus, the operating wavelength range in our photodetector can be extended with a higher bias. Phonon involved excitation processes and the energy diagrams with the applied bias voltages are shown in Figure 1. Once the phonon-assisted electron–hole pairs are produced, the number of carriers is amplified through the avalanche multiplication. Hence our device shows high photoresponse characteristics for a wide spectrum of light, overcoming the limitations of conventional photodetectors. Consequently, our device outperforms with the widest spectral range from violet (405 nm) to near-infrared (1310 nm) and highest responsivity over $10^4$ A W$^{-1}$ compared to other 2D-material-based APDs. These results suggest significant potential of bandgap-independent broad-spectrum photodetection of 2D layered materials through efficient carrier multiplication.

2. Results and Discussions

The upper panel of Figure 1a shows a schematic of a WSe$_2$ APD under illumination. Mechanically exfoliated WSe$_2$ ($\sim 12$ nm) was transferred onto a 285 nm SiO$_2$/Si substrate, and Cr/Au electrodes for source and drain were deposited onto WSe$_2$ (see the Experimental Section and Section S1 (Supporting Information) for a detailed description of the fabrication and characterization of the WSe$_2$ flake and device). All measurements were performed under ambient conditions at room temperature. We applied a drain voltage from 0 to $\sim 50$ V, corresponding to $E = 0$–100 kV cm$^{-1}$ (channel length: 5 μm), at a fixed $V_{GS} = -20$ V, which is the charge neutral point. We set $V_{GS}$ as the charge-neutral point to secure the lowest dark current (see Section S2a, Supporting Information). As the electric field increased, the channel current increased owing to an increase in the number of injected carriers from the metal electrode by thermionic emission at a low electric field and saturated to some extent at $I_{sat}$ (saturation current). Subsequently, it increased exponentially by avalanche multiplication in the high-bias regime over $V_{BR}$ (i.e., avalanche breakdown voltage). As shown in Figure 1b, we can observe the characteristic of avalanche multiplication in which the channel current increases exponentially after saturation in both log scale (black line) and...
linear scale (blue line). Thus, we can set avalanche breakdown voltage \( V_{BR} \) where avalanche multiplication initiated as \(-17.5 \text{ V}\). Figure 1c shows the dark current (black line) without any introduced light and the corresponding multiplication factor (blue circles), defined as \( M = \frac{I_{DS}}{I_{sat}} \). (Here, \( I_{sat} = \approx 0.2 \text{ nA at } V_{BR} = -17.5 \text{ V} \)). WSe\(_2\) shows a multiplication factor of over \( 10^4 \), which is a record-high value indicating that a large amount of carrier multiplication process occurs in WSe\(_2\) channel. The injected holes accelerated sufficiently under a high electric field and collided with the lattice with high kinetic energy to generate free carriers. These generated free carriers accelerated and repeated the process of generating another free carrier, causing an abrupt current increase in the WSe\(_2\) channel. The red line in Figure 1c shows the channel current measured under light with a wavelength of 1310 nm and an intensity of 500 nW cm\(^{-2}\). This illumination current is twice as high as the dark current at high electric field, which is a result of light-excited carriers and their carrier multiplication via the avalanche effect. As shown in the lower panel of Figure 1a, photoinduced carriers undergo the avalanche multiplication process in the same way as the injected carriers when subjected to a high electric field. The difference between the illumination and dark current is defined as \( I_{ph} \) (photocurrent), which refers to the effect of the irradiated light on the channel. Figure 1d shows the photocurrent (blue circles) and the corresponding responsivity (black circles). This photocurrent also increases slightly after \( V_{BR} \) and eventually increases exponentially, which means that the photogenerated carriers also undergo avalanche multiplication under a high electric field. Because of the high photocurrent resulting from this enormous carrier multiplication process, the responsivity of the WSe\(_2\) APD was as high as \( 10^7 \text{ A W}^{-1}\). Responsivity is defined as the ratio of the output photocurrent to the input light power and is calculated by

\[
R = \frac{I_{ph}}{P}
\]

where \( P \) is the incident power. This high value of \( R \) (responsivity) indicates the generation of numerous carriers by the incident light.

The detailed photoresponse characteristics of the WSe\(_2\) APD are shown in Figure 2 with the incident light wavelength of 1310 nm. The channel current under light illumination (illumination current) and the photocurrent under different incident power densities are shown in Figure 2a,b, respectively. As shown in Figure 2a, the illumination current increases as the incident power increases. This is because the number of photogenerated carriers increases as the intensity of light increases, and additional free carriers, multiplied by them under a high electric field, increase. This phenomenon corresponds to the increasing trend of photocurrent, as shown in Figure 2b. Figure 2c shows the light power density dependent photoresponse characteristics. The detectivity \( (D^*) \) of the photodetector can be defined as

\[
D^* = \left( \frac{A \Delta f}{NEP} \right)^{1/2}
\]
where $A$ is the device area, $\Delta f$ is the bandwidth, and $\text{NEP}$ is the noise-equivalent power. This can be derived as

$$D^* = \frac{A}{2eI_{dark}M'}F(M)$$

where $e$ is the elementary charge, $M$ is the multiplication factor, $F(M) = kM + (1 - k)(2 - 1/M)$, and $k$ is the ratio of the electron and hole ionization coefficients ($0 < k < 1$). See Section S3a (Supporting Information) for detailed derivation and calculation.

Both the responsivity and detectivity decrease as the incident power density is increased, with a similar dependence to previously reported photodetectors, but they are maintained over $10^4$ A W$^{-1}$ and $10^7$ Jones, respectively.

Such ultrasensitive photoresponse characteristics can be attributed to a high-gain mechanism that allows one incident photon to induce multiple carriers to conduct a current in the device. As discussed, the superior avalanche multiplication in WSe$_2$ acts as the high-gain mechanism in our device with a record-high avalanche gain of $\approx 10^5$, as shown in Figure 2d. We obtain the avalanche gain$^{[15,33]}$ by

$$\text{Avalanche gain} = \frac{I_{ph}}{I_{ph0}}$$

where $I_{ph0}$ is the photocurrent immediately before avalanche multiplication. Here, $I_{ph0}$ was considered at 35 kV cm$^{-1}$, which is the critical electric field of WSe$_2$. The avalanche gain indicates the efficiency of carrier multiplication by incident photons, and this power-independent high value implies the occurrence of a large amount of carrier multiplication via the avalanche effect of WSe$_2$.

In our WSe$_2$ APD, a high photoresponse was observed in the broad spectrum from violet (405 nm) to near-infrared (1310 nm) wavelengths. Figure 3a shows the photocurrent as a function of the applied electric field at various wavelengths. The power density of all incident lights was fixed at 500 nW cm$^{-2}$. A high photocurrent of over 60 µA observed with light illumination of 405 nm wavelength, and a photocurrent of 4 µA was confirmed with 1310 nm wavelength light. The previously reported conventional WSe$_2$ photodetectors showed low photoresponse characteristics to light with wavelengths over 850 nm.$^{[21–27]}$ However, our WSe$_2$ APD overcomes this problem under a high E-field owing to phonon-assisted photogeneration.

The influence of an electric field on the optical absorption of a semiconductor in the vicinity of an absorption edge has been previously studied.$^{[28–34]}$ It was found that absorption occurs for photon energies lower than the ordinary bandgap. The optical absorption in crystals caused by the interband transitions of electrons is altered in the presence of an external electric field (electro-absorption effect), and the phonon-assisted indirect transitions increase with the field. In the absence of an electric field, the absorption coefficient, $\alpha(\omega)$, was considered by Elliot.$^{[35]}$ This theory is applicable to direct bandgap transitions. In the presence of a field, phonon-assisted indirect transitions

![Figure 2. (a) Illumination current and b) photocurrent response of the WSe$_2$ APD under different light illumination power densities. The illumination wavelength is 1310 nm. Each dashed line represents a trend line, and enlarged filled circles represent values under an E-field of 100 kV cm$^{-1}$. c) Variation in responsivity (black circles) and detectivity (blue squares) as a function of incident power. d) Corresponding avalanche gains upon various incident power. Record-high avalanche gain of $\approx 10^5$ indicates that photogenerated carriers keep performing avalanche multiplication well under high electric field, despite of very small power intensity.](image-url)
contribute to optical transitions, and the absorption coefficient for these indirect processes has been considered. However, no exact expression for the absorption coefficient is available, and approximate expressions for \( \alpha(\omega) \) near the band edge are provided. According to the above references, the change in the absorption coefficient upon the application of an electric field \( \Delta \alpha(F) = \alpha(F) - \alpha(0) \), is given by

\[
\Delta \alpha(F) = \beta F^{4.5} \left( \frac{\hbar \omega - E_{ph}}{\hbar \omega + E_{ph}} \right)^{1/2} \left( 1 - \frac{1}{4 \sqrt{\pi}} \xi^{5/2} \cos \frac{4}{3} \xi \right) \tag{5}
\]

where \( \beta \) and \( \gamma \) are field-independent numerical constants that contain the fundamental materials and measurement parameters, respectively. \( \xi \) is a function of the electric field, \( F \), which is given as \( \xi(F) = \left( \frac{2 \mu}{\hbar^2 c^2} F \right)^{1/3} \left[ \hbar \omega - E_{ph} + \hbar \omega_{ph}(q) \right] \) with the reduced mass \( \mu \); \( \hbar \omega \) and \( \hbar \omega_{ph}(q) = E_{ph} \) are the incident photon energy and the energy of the phonon responsible for momentum conservation in the process, respectively.

In Figure 3b, we show the processes of the phonon-assisted optical transition in the presence of a bias (an electric field). The process A indicates the direct optical interband transition. Since bulk WSe\(_2\) has an indirect bandgap, the incident photon energy \( (\hbar \omega) \) must be higher than the indirect bandgap \( (E_{ig}) \) of WSe\(_2\) (i.e., \( \hbar \omega > E_{ig} \)) to produce an electron–hole pair. The processes B and C indicate the phonon-assisted optical interband transition. In process B, the valence-band electron first absorbs an incident photon to reach a virtual state, then absorbs or emits a phonon to reach a final state in the conduction band. In process C, the valence-band electron first absorbs or emits a phonon and then absorbs a photon to reach the conduction band. In the final state, there is an electron in the conduction band and a hole in the valence band state, and a phonon has been either created or annihilated. In these phonon-assisted optical absorptions, the required incident photon energy is given by \( \hbar \omega > E_{ig} + \hbar \omega_{ph} - \alpha \) for a phonon absorption (+) and emission (−), where \( E_{ph} \) is the phonon energy and \( \alpha \) is a parameter given by the ratio of the phonon wavelength and the sample size. This parameter arises from the momentum conservation in the processes, and we estimate \( \alpha \approx 0.01 \) in our device because the order of the dominant phonon wavelength is order of nm at room temperature. The detectable wavelength, determined by the energy bandgap of WSe\(_2\), is less than 1240 nm (here, the energy bandgap of bulk WSe\(_2\) is estimated to be 1.0 eV). However, under the influence of the phonon-assisted process with a drain voltage over \( V_{BR} \), the detectable wavelength range can be extended to 1550 nm, which corresponds to 0.8 eV of energy bandgap. We note that in our WSe\(_2\).
APD, the incident photon energy for the pair production can be further reduced with a higher drain voltage, and the detection of longer wavelength light can be possible. UV and visible light with photon energies beyond the bandgap can be detected even before avalanche multiplication occurs through a conventional mechanism, as shown in the left panel of Figure 3b.

At low bias voltage, before avalanche multiplication occurs, our APD has low photoresponse for long-wavelength light, similar to conventional WSe$_2$ photodetectors. However, at high enough bias voltages to stimulate the avalanche multiplication, the carriers produced by the phonon-assisted optical absorption are accelerated under a high E-field, and their number is amplified through the impact ionization process. It leads to a high responsivity, of over $10^7$ A W$^{-1}$ for broad-spectrum, of our WSe$_2$ APD (405–1310 nm) light under a high E-field, as shown in Figure 3c. As shown in Figure 3c, for the region of $E < E_{CR}$ (dotted black line), WSe$_2$ cannot detect long-wavelength light with a similar tendency to the previously reported WSe$_2$ photodetectors (colored circles in Figure 3d). Relatively high responsivity in the low electric field regime has been achieved through complicated processes such as chemical doping$^{[24,25]}$ and contact engineering.$^{[23,26]}$ Whereas higher responsivity can be achieved by simply increasing the electric field over the $E_{CR}$, as shown by the red square. When the electric field exceeds the $E_{CR}$, high-gain carrier multiplication originating from the avalanche effect of the WSe$_2$ channel acts on the photoinduced carriers. These high responsivities for the broad spectrum from 405 to 1310 nm are nearly independent of the wavelength of the incident light.

From the relation between the responsivity and dark current, we calculated the normalized photocurrent-to-dark current ratio (NPDR), which is defined as

$$NPDR = \frac{R}{I_{dark}}$$  \hspace{1cm} (6)

where $R$ is the responsivity and $I_{dark}$ is the dark current. The WSe$_2$ APD operated well for a broad light spectrum with an NPDR of $1.6 \times 10^{14}$ W$^{-1}$ under 405 nm light illumination and $5.7 \times 10^{12}$ W$^{-1}$ under 1310 nm light illumination. In Figure 4, we compare the spectral NPDR and responsivity of our device to other 2D layered-material-based APDs. Detailed parameters are listed in Table S1 (Supporting Information). This benchmark graph shows that this work not only has high responsivity and NPDR values compared to APDs using other 2D layered materials but also has the ability to cover a broad spectrum from violet (405 nm) to near-infrared (1310 nm).

3. Conclusions

In conclusion, we demonstrated broad-spectrum photodetection with high sensitivity under a high E-field using WSe$_2$ APD. The main operating mechanism of our device is high-gain carrier multiplication by impact ionization and phonon-assisted photogeneration. WSe$_2$ outperformed other 2D layered materials in terms of impact ionization characteristics with a record-low critical E-field of 35 kV cm$^{-1}$ and a high multiplication factor of over $10^4$. Owing to its superior avalanche multiplication, the photogenerated carriers were also multiplied by a high-gain mechanism under a high E-field, the device exhibited a high photosresponsivity of $10^9$ A W$^{-1}$. We reported high responsivities of over $10^7$ A W$^{-1}$ for a wide spectral range from violet (405 nm) to near-infrared (1310 nm). Hence, the proposed WSe$_2$ APD outperforms not only previously reported 2D layered-material-based APDs but also other WSe$_2$ based photodetectors. Our results suggest a promising general approach for developing a single photodetector that can cover a broad spectrum of light with a high sensitivity for future optical communication.

4. Experimental Section

Device Fabrication: 1.2 × 1.2 cm$^2$ SiO$_2$ (285 nm)/p$^+$-doped Si substrates were cleaned by acetone and IPA with ultrasonification for 10 min and dried with N$_2$. WSe$_2$ flakes were obtained via mechanical exfoliation onto a substrate from bulk WSe$_2$ using the blue tape (2245PV, Nitto) exfoliation method. The tape residue was removed by placing the samples in an acetone bath at 120 °C for 30 min. It was coated with PMMA (495 A4, 950 A9) using a spin coater at a rotation speed of 4000 rpm for 60 s. E-beam lithography was used to fabricate backgate FETs, and a Cr/Au (5/50 nm) electrode was deposited using an e-beam evaporator with 0.1 Å s$^{-1}$ (Cr), 1.0 Å s$^{-1}$ (Au), followed by a lift-off process. See Figure S1 in the Supporting Information for a schematic illustration of the process flow.

Characterization: The structures and surface morphologies of the flakes and devices were measured using optical microscopy (BX 51 M, Olympus). Using a 532 nm excitation wavelength laser, the Raman spectra were measured using a laser micro-Raman spectrometer (Kaiser Optical Systems RXN). The thickness was confirmed by AFM measurements (Park Systems Corp.) using a noncontact mode with PPP-NCHR probe tips (nanosensors). A Keithley 4200 parameter analyzer was used to perform all electrical measurements under ambient conditions at room temperature. The photoresponse of the phototransistor devices was measured using a Keithley 4200 parameter analyzer equipped with a vertically aligned monochromatic laser illumination system at various power densities.

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

2D WSe₂, avalanche photodetector, broad-spectrum photodetector

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