Approaching the Intrinsic Threshold Breakdown Voltage and Ultrahigh Gain in a Graphite/InSe Schottky Photodetector

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Realizing both ultralow breakdown voltage and ultrahigh gain is one of the major challenges in the development of high-performance avalanche photodetector. Here, it is reported that an ultrahigh avalanche gain of $3 \times 10^5$ can be realized in the graphite/InSe Schottky photodetector at a breakdown voltage down to 5.5 V. Remarkably, the threshold breakdown voltage can be further reduced down to 1.8 V by raising the operating temperature, approaching the theoretical limit of $1.5 E_g/e$, with $E_g$ the bandgap of semiconductor. A 2D impact ionization model is developed and it is uncovered that observation of high gain at low breakdown voltage arises from reduced dimensionality of electron–phonon scattering in the layered InSe flake. These findings open up a promising avenue for developing novel weak-light detectors with low energy consumption and high sensitivity.

1. Introduction

Avalanche photodetectors (APDs) enable the detection of ultralow light levels via charge amplification mechanism and have been widely used in many fields including weak light detection.[1–3] 

2. Results and Discussion

As schematically shown in Figure 1a, vdW Schottky APD was fabricated based on graphite/InSe vdW heterostructures,
which were assembled by dry transfer technique. Here, InSe has an ultrahigh in-plane mobility (≈10^4 cm^2 V^−1 s^−1 at 4 K and ≈4000 cm^2 V^−1 s^−1 at room temperature), which ensures a rapid acceleration of carriers to facilitate the impact ionization process. In addition, multilayer InSe possesses a direct bandgap of ≈1.2 eV and is promising to achieve a high quantum efficiency. Meanwhile, by employing graphite flake as the vdW Schottky contact, we can achieve an atomically flat interface and thus avoid the interface quality degradation which is inevitable in conventional metal contact. Such high interface quality can significantly suppress the dark current and thus is critical to achieve high gain.

InSe flake (30–50 nm) onto highly p-doped silicon substrates covered by 300nm SiO_2, and graphite flakes (5–20 nm) onto a poly(dimethylsiloxane) (PDMS) sheet. Then the graphite flakes were transferred on top of InSe flakes to construct the graphite/InSe Schottky junction. Standard e-beam lithography followed by e-beam evaporation was employed to fabricate 5 nm Ti/45 nm Au electrodes. The gate voltage V_{gs} was applied to deplete the free charge carriers, thus providing a wide depletion region to facilitate the avalanche breakdown (see Figure S1, Supporting Information). Note that the electrical transport measurements were performed with the graphite/InSe Schottky junction reverse-biased and InSe/Ti Schottky junction forward-biased, unless specified otherwise.

We first studied the current variation (I_{ds}) and multiplication as a function of drain–source voltage (V_{ds}) in a typical device, with the results shown in Figure 1b. The current measured at the dark condition is on the order of picoampere at low V_{ds} and is at least six orders of magnitude smaller than the dark current in conventional APDs, which demonstrates the benefit of vdW Schottky contact. Remarkably, the measured current exhibits an abrupt increase of more than five orders above a certain threshold voltage, that is, V_{bd} = 5.5 V, suggesting that an avalanche breakdown occurs. It should be noted that the graphite/InSe APD can exhibit avalanche breakdown even at room temperature (see Figure S2, Supporting Information). By illuminating the InSe flake by a laser with a wavelength of 532 nm at 6.9 pW, we observed a sharp rise in the photocurrent at a smaller threshold voltage, that is, V_{bd} = 5.5 V, suggesting that an avalanche breakdown occurs. It should be noted that the graphite/InSe APD can exhibit avalanche breakdown even at room temperature (see Figure S2, Supporting Information).
the light responsivity and quantum efficiency of our device were measured to be \(1.16 \times 10^5\) A W\(^{-1}\) and \(2.7 \times 10^5\), respectively, indicating that graphite/InSe Schottky photodetector exhibits a high sensitivity and thus facilitates the detection of weak light signals. Compared to traditional APDs\(^{[7,8,19]}\), our devices exhibit three orders of magnitude higher avalanche gain, and five orders of magnitude lower dark current at the same breakdown voltage. These advantages highlight the great potential of our graphite/InSe APDs in practical applications requiring low energy consumption and high sensitivity. As the laser power \(P_{\text{opt}}\) is raised (Figure 1c,d), the measured photocurrent as a function of reverse bias voltage exhibits two distinct features. Within the regime of low \(V_{\text{ds}}\), the photocurrent gradually increases with the light intensity. Such an increase in photocurrent can be attributed to the photoconductive effect that the electrical conductivity is enhanced by the photogenerated free electrons and holes in InSe. Meanwhile, in the regime that \(V_{\text{ds}}\) is above the breakdown voltage and the laser power is low, that is, below 6.9 µW, the photocurrent is almost unchanged as we increase the light intensity, as shown in Figure 1c,d. We attribute the unconventional \(I–V\) characteristics under different light illumination to the limitation of series resistance.\(^{[20–22]}\)

The experimental result of such high gain up to \(10^5\) at a low breakdown voltage cannot be explained by the traditional model of avalanche breakdown. In the conventional APDs, the charge multiplication mechanism is based on a one-carrier cascade ionization process, and therefore, an excessively high breakdown voltage of at least 30 V is required for achieving a gain of \(10^5\) (see Section S4, Supporting Information), while it can be accessed at \(V_{\text{bd}} = 5.1\) V in our observations. We also carefully checked the channel length dependence of the breakdown voltage in the InSe-based APDs, showing similar breakdown voltages for different channel lengths (see Figure S4, Supporting Information), which is also inconsistent with the model of one-carrier impact ionization.\(^{[23]}\)

We attribute our observation to the quantum confinement effect induced by the vdW gap in the layered InSe, which facilitates an alternative mechanism, that is, a two-carrier process.\(^{[24,25]}\) As shown in Figure 2a, the vdW gap acts as a tunneling barrier of \(\approx 1.85\) eV (see Figure S5, Supporting Information) and suppresses the out-of-plane charge transport,\(^{[26,27]}\) leading to the reduction in the dimensionality of e–ph scattering and the enhancement of the Coulomb interaction.\(^{[12,14]}\) In this sense, the energy dissipation via e–ph scattering will be suppressed, and the impact ionization rate will be enhanced,\(^{[14]}\) enabling the two-carrier impact ionization process which can explain the observation of a much higher gain at a much lower breakdown voltage compared to previous works (see Figure S22, Supporting Information). Specifically, as schematically shown in Figure 2b, when an electron is injected into the vdW Schottky junction and accelerated under the action of both external and built-in electrical field to initialize an impact ionization event, an electron-hole pair will be generated. Then the generated hole will drift back to produce another electron-hole pair and the two

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**Figure 2.** a) Impact of reduced dimensionality of e–ph scattering on the electron acceleration process. b) Illustration of the two-carrier impact ionization process. c) Ionization rate \(\alpha_{2D}\) and d) Gain as functions of electric field strength \(E\) and mean-free path length \(\lambda\). The black dashed line indicates that impact ionization probability \(p = \alpha \times \lambda\) is equal to 1 and gain \(M\) is divergent. e) \(\alpha\) and \(p\) as functions of field strength \(E\) in two dimensions (red solid line) and three dimensions (blue solid line). The black dashed lines and blue dashed lines are the asymptotes of \(\alpha_{2D}\) and \(\alpha_{3D}\), respectively. The horizontal black dotted line corresponds to the case where \(p = 1\). f) Gain as functions of \(E\) in two dimensions (red solid line) and three dimensions (blue solid line). The vertical gray dashed line corresponds to the case where gain \(M\) is divergent.
electrons are collected by the electrode. As the cycle repeats, more and more carriers are generated in a region within the length of only one mean-free path \( \lambda \) and collected through electrodes, which explains the channel length independence of breakdown voltage in our device. Assuming electrons and holes have equal ionization probability \( p \), the avalanche gain for the two-carrier impact ionization process can be determined by \( M = p/(1-p) \).\(^{24}\) Accordingly, if \( p \) approaches 1 due to the reduction in the dimensionality of e–ph scattering, ultrahigh avalanche gain can be reached at an extremely low breakdown voltage, which is consistent with our results.

To further investigate the ionization process in layered InSe, we carried out theoretical calculations considering the effect of reduced dimensionality of e–ph scattering. Note that the ionization probability \( p \) is usually expressed by the ionization rate per unit path length \( \alpha \) (i.e., \( p = \alpha \times \lambda \)), which is an intrinsic property of semiconducting material characterizing the multiplication process.\(^{28}\) Assuming that the carrier transport in InSe is strongly confined within the 2D atomic layers while the carrier transfer between adjacent layers can be neglected, we find that the 2D ionization rate \( \alpha_{2D} \) can be described by

\[
\alpha_{2D} \propto \frac{E}{E_{k,th}}
\]

Here \( E \) is the electric field strength, and \( E_{k,th} \) is the threshold carrier kinetic energy \( E_{k,th} = 1.5E_g \) above which the charge multiplication occurs. Here we adopted the methods developed by Wannier\(^{29}\) and Wolff\(^{28}\) for solving the Boltzmann equation in the high-field limit, and generalized these methods to a 2D geometry (see Section S8, Supporting Information). Assuming there is negligible energy loss to phonons per collision, we only need to solve for the zeroth and first-order terms of the carrier velocity distributions, which in turn allow us to derive an expression for the 2D ionization rate \( \alpha_{2D} \). In the limit where the kinetic energy gained by the carrier accelerated over \( \lambda \) by the field \( E \) far exceeds the energy lost to phonons (i.e., \( h\omega \ll eEd\lambda \)), we arrive at Equation (1) above.

Based on this developed model, we theoretically examine mean-free path \( \lambda \) and electrical field \( E \) dependence of the ionization rate \( \alpha_{2D} \) and gain \( M \), with results shown in Figure 2c,d, respectively. The gain \( M \) shown in Figure 2d is given by multiplying the gain obtained from a single injected electron by the layer number (=40) of the InSe flake used in our experiment. Note that there exists a white region in Figure 2d where \( \alpha_{2D} \) cannot be simultaneously achieved. Although the divergent \( M \) cannot be accessed in our experiment due to the limitation of resistance, it is in good agreement with the ultrahigh gain observed in our experiments.

To further demonstrate that the reduced dimensionality of e–ph scattering indeed plays a significant role in the ionization rate, we compare \( \alpha_{2D} \) with the ionization rate \( \alpha_{3D} \) for bulk geometries whose carrier motion is isotropic in three dimensions. By fixing \( \lambda = 22 \) nm which is the value extracted from our experiments (see Section S7, Supporting Information), we obtain \( \alpha_d \) (where \( d \) denotes either 2D or 3D) and the gain \( M \) obtained from a single injected electron as functions of \( E \), as shown in Figure 2e,f. We find that \( \alpha_{2D} \) is asymptotically described by Equation (1) and reaches \( p = 1 \) (black dotted line) at high fields, as shown in Figure 2e. In contrast, at large \( E \), \( \alpha_{3D} \) saturates to \( C/\lambda \) with \( C = \sqrt{3}d/2 \) (see Equation S73, Supporting Information), indicating a \( p = \lambda/\alpha_{3D} \) smaller than 1 for all \( E \), which inherently limits the possible increase in gain with a corresponding increase in \( E \) (Figure 2f). The divergent gain for the 2D case and the saturation for the 3D case in turn support the foregoing hypothesis that the ultrahigh gain observed in our vdW device results from the reduced dimensionality of the e–ph scattering in the layered material.

According to our model, the suppressed energy loss to lattice and the increased ionization rate in the layered InSe allow for the realization of intrinsic threshold breakdown voltage approaching the theoretical limit of 1.5 \( E_g/e \) accompanied with a high gain. To search for this intrinsic threshold breakdown voltage, we varied the temperature to continuously tune the lattice vibration, which can modify e–ph scattering strength and thus the breakdown voltage.\(^{30}\) Figure 3a plots the drain–source current \( I_d \), as a function of bias voltage \( V_{ds} \) at different temperatures ranging from 140 K to 260 K. Remarkably, we observed a breakdown voltage as low as 1.8 V at \( T = 260 \) K, which approaches the theoretical limit of threshold breakdown voltage 1.5 \( E_g/e \) by using the bandgap (i.e., \( E_g = 1.2 \) eV) of InSe. This is the first time that an intrinsic threshold breakdown voltage has been observed experimentally. We attribute this to the outstanding conductivity of the graphite contact, which makes the bias voltage drop mainly on InSe and thus facilitates ionization. At this breakdown voltage, we achieved an avalanche gain \( \approx 110 \). It is worth noting that a breakdown voltage of at least 12 V is required for conventional materials to access such a high gain (see Section S4, Supporting Information), indicating that the use of vdW material gives an unprecedented opportunity for addressing the traditional challenge that the intrinsic threshold breakdown voltage and the high avalanche gain cannot be simultaneously achieved.

We now turn to the temperature dependence of the breakdown voltage and the avalanche gain. As shown in Figure 3b, the breakdown voltage monotonically decreases with the rising temperature while the gain exhibits non-monotonic temperature dependence, both of which are fundamentally different from the features widely reported in conventional APDs. To interpret such unusual temperature dependence of the breakdown voltage and the gain, we investigate three consecutive electron transport processes, that is, I) injection, II) ionization, and III) collection,\(^{31}\) as schematically shown in the inset of Figure 3a. First, at high temperatures, the ionization process is suppressed since the mean-free path is reduced due to enhanced lattice vibrations. In this case, a larger breakdown voltage is required to provide a stronger electrical field to accelerate electrons to obtain the threshold energy, which is inconsistent with the negative temperature coefficient of \( V_{bd} \) shown in Figure 3b. In contrast, in the injection and collection processes, as electrons possess more thermal energy with the increasing temperature, the electric field strength required to pass over the potential barrier decreases. Moreover, the number of injected electrons in the injection process are much less than that in the collection process, indicating a negligible temperature effect in the injection process. Thereby, we can attribute
the negative temperature coefficient of breakdown voltage to the temperature-enhanced collection process through InSe/Ti Schottky junction. Different from the collection-dictated monotonical temperature dependence of the breakdown voltage, the non-monotonical temperature coefficient of the gain is governed by the competition between ionization and collection. As the temperature rises from 80 K to 180 K, the gain is limited by the collection efficiency, which is consistent with the experimental result shown in Figure 3a that the reverse saturation current increases as temperature increases. As the temperature is further raised, that is, from 180 K to 260 K, the mean-free path is significantly reduced and the ionization process becomes the major limiting factor, despite the collection efficiency continues to increase.

The underlying physics described above can also be justified by reverse-biasing the InSe/Ti Schottky junction, which leads to a positive temperature dependence of the breakdown voltage ranging from 80 K to 180 K and a decrease in gain from the lower temperature of 100 K, as shown in Figure 3c,d. Note that the InSe/graphite Schottky barrier height is much smaller than that in the InSe/Ti junction, as supported by the thermionic emission measurement shown in Figure S16 (Supporting information), making the ionization generated electrons pass through the InSe/graphite barrier, that is, the collector terminal, much easier. In this case, the impact ionization would be the major factor for determining the breakdown voltage and the gain, and thus leads to a positive temperature dependence of breakdown voltage and a decrease in gain from a lower temperature, as we observed. To further systematically investigate the temperature coefficients for different Schottky barrier heights, we have designed three sets of control experiments, with the result confirming that the sign of temperature coefficient is a result of the competition between the collection and ionization processes (see Figure S15, Supporting Information).

### 3. Conclusion

We have observed an intrinsic threshold breakdown voltage of 1.8 V as well as ultrahigh avalanche gain $\approx 3 \times 10^5$ in the graphite/InSe Schottky APD. Based on a 2D carrier multiplication model, we attribute the results to the emergence of a two-carrier ionization process that is facilitated by the reduced dimensionality of e–ph scattering in layered InSe semiconductors. In addition, we demonstrate that both the breakdown voltage and avalanche gain not only depend on temperature-dependent ionization but are also affected by the thermally assisted collection process. These findings reveal
the fundamental difference in carrier multiplication physics between conventional and layered semiconductors and open up new perspectives for future APDs with both low energy consumption and high sensitivity.

4. Experimental Section

Device Fabrication and Measurement Setup: First, the InSe flakes (30–50 nm) were mechanically exfoliated onto highly p-doped silicon substrates covered by 300 nm SiO2, and graphite flakes (5–20 nm) onto the PDMS sheet. Then the graphite flakes were transferred on top of InSe flakes to construct the graphite/InSe Schottky junction. Standard e-beam lithography followed by e-beam evaporation was employed to fabricate 5 nm Ti/45 nm Au electrodes. The Ids–Vds curves of the Ti/InSe/graphite were measured when the gate voltage Vgs = −40 V (Figures 1 and 3). All the electrical measurements were performed by using a Keithley 2636B dual-channel digital source meter. The SAPPHIRE 532-20 CW SF CDRH laser was used as the light source, the spot size of which was focused to 1 μm in diameter. The change of temperature in the electric measurements was performed with an Oxford instrument Microstate HiRes.

Characterizations: The thickness of the sample was determined by the atomic force microscope using Bruker MultiMode 8. The Raman spectra of the sample were acquired by using WiTec alpha300 under 532 nm excitation. The transfer curve of InSe and the Ids–Vds curve of the Ti/InSe/graphite were acquired using a Keithley 2636B source meter.

Calculations of Ionization Rate for 2D Material: First, the methods developed by Wannier[29] and Wolff[30] were adopted for solving the Boltzmann equation in the high-field limit, and generalized to a 2D geometry. Then the approach undertaken by Wolff was used to solve for the velocity distributions in the 2D case. Finally, by solving for the velocity distribution, it could obtain the expressions for the 2D ionization rate per unit length as well as the gain M (Figure 2c–f) (see Sections S8 and S9, Supporting Information).

Statistical Analysis: The data was used without any transformation, except for the following: Figure 1b, Figure 3b, and Figure 3d. Figure 1b presents the multiplication factor M based on McIntyre’s formula (M = (Iph − Igs)/Igs with Igs the photocurrent at M = 1).[31] In Figure 3, Igs–Vds characteristic of the same Ti/InSe/graphite device was performed for two cycles at the dark condition at a different temperature, where a single cycle is presented in Figure 3a and Figure 3c. In Figure 3b and Figure 3d, the gain and breakdown voltage were given in the form of mean ± standard deviation. The statistical analysis was carried out using IGOR Pro.

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

avalanche photodiodes, InSe, layered materials, van der Waals (vdW) Schottky junctions

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