

Tunneling-barrier-controlled sensitive deep ultraviolet photodetectors based on van der Waals Heterostructures

Corresponding Author: Professor Jianlu Wang

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors investigated new photodetection scheme based on tunneling barrier modulation rather than the band gap of the absorber material, and achieved highly sensitive DUV photodetection using the two-dimensional van der Waals heterostructures. This is interesting research.

How does the performance change with varying hBN thicknesses?

How does defects at the interface of MoS₂/hBN/FLG affect the photodetector performance?

Reviewer #2

(Remarks to the Author)

Li et al report an interesting result on tunneling barrier controlled ultrasensitive deep ultraviolet photodetector based on van der Waals heterostructures, demonstrating application potential for solar-blind detection application. The authors have developed a complex device structure and operating process that enhances the optical gain of traditional devices, however, the reported response time of 0.5 s is significantly longer than what is typically expected for tunneling-type electronic devices, which are known for their high-speed response. This long response time undermines the advantages of the proposed device and is difficult to accept. several critical concerns lead me to recommend rejection of the manuscript:

1. The two-dimensional materials generally pose challenges for ultraviolet light absorption and photoelectric conversion efficiency. Therefore, using two-dimensional materials as the light absorption layer does not seem optimal. Although the cutoff response wavelength in tunneling devices is determined solely by the barrier height of the heterojunction (independent of material bandgap), selecting wide bandgap semiconductors with high absorption coefficients for the absorption layer would better facilitate efficient photoelectron generation and improve the optical gain of UV detectors.
2. The design where the incident light enters from the top while the photosensitive absorption layer is positioned beneath the transport layer is unusual for photodetector structures. An explanation for this design choice is necessary.
3. Without tunneling involvement, it remains unclear how to differentiate between the photogenerated current in the transport layer and the contribution from tunneling to the final photocurrent gain.
4. In line 162 of the manuscript, please provide specific values for the uncommon parameters m_b and m_s used in the equation.
5. Is a gate voltage necessary to drive the photogenerated electrons/holes from the absorption layer to the transport layer? In Figure 2g, it is stated that under UV illumination, photogenerated electrons in the absorption layer tunnel into the transport layer, resulting in an extremely high photocurrent. However, without gate voltage assisting the tunneling process, clarification is needed regarding the driving force for the electrons to cross the barrier and inject into the transport layer.
6. Supporting Data for Figure S7: Providing time-resolved photoresponse data for VFLG at 0 V and 3V conditions would add more persuasive evidence. Additionally, does VFLG>0 favor obtaining a larger optical gain?
7. The manuscript needs thorough language proofreading to ensure accuracy and fluency in academic writing, as well as adherence to the journal's formatting guidelines.
8. Regarding Fig. 15 in the supporting material, the current at $V_d = 0.1$ V is only an order of magnitude lower than at 0 V with frequency more than 1000 Hz, which is quite unusual. Moreover, comparing it with the dark current data in Fig. 2h of the main text, the current density may have been underestimated.
9. It is unreasonable to simply compare Figure 2g and Figure 7 in the supporting materials to demonstrate the advantages of the ultra-high photoresponse of tunneling devices. They do not operate in similar modes and lack reasonable controlled

variables. Figure 2g is a floating gate device, while Figure 7 in the supporting materials operates similarly to a transistor. The photocurrent generated by the absorption layers, such as graphene, seems unlikely to produce such a large photocurrent as a source of tunneling current; please provide the response curves of graphene and other light-absorbing materials under violet light.

10. In summary, the operational mode of this structure is more akin to a memory that is electrically written and optically erased. This structure is better suited to utilize the principles of optical floating gate transistors for memory instead of detectors.

Reviewer #3

(Remarks to the Author)

What are the noteworthy results?

The authors have presented deep ultraviolet (DUV) photodetection with impressive performance metrics (e.g., responsivity $\sim 4 \times 10^4$, $I_{on}/I_{off} \sim 106$) using MoS₂ as the charge transport layer and few-layered graphene (FLG) as the photon absorption layer, separated by a hexagonal boron nitride barrier layer. The authors claim that the novelty lies in the DUV photodetector's cut-off wavelength tuneability through an electrically modulated electron F-N tunnelling barrier, which is not limited to the bandgap of the absorption material. Please refer to the comments below.

- Will the work be of significance to the field and related fields?

Yes, because the author has presented results on deep UV detectors with outstanding performance metrics.

How does it compare to the established literature? If the work is not original, please provide relevant references.

The work is original. However, the device architecture presented in this work has similarities with the other reports (DOIs mentioned below):

doi: 10.1021/acs.nanolett.6b04449, 17, 453–459

doi: 10.1109/LPT.2020.2993444

- Does the work support the conclusions and claims, or is additional evidence needed?

The author has supported their claims with data; however, additional evidence is needed. Please refer to my other comments to know more about the required revisions.

- Are there any flaws in the data analysis, interpretation, and conclusions? - Do these prohibit publication or require revision?

To support their claim, the author should revise the manuscripts with some additional evidence, such as hBN's (barrier layer) deep UV absorption, charge carrier generation, etc. Please refer to my other comments below to learn more about the required revisions.

- Is the methodology sound? Does the work meet the expected standards in your field?

Yes, the methodology is sound, and the author has supported the claim with relevant data; however, in a few areas, it still needs clarity before publication. Please refer to my other comments below to know more about the required revisions.

- Is there enough detail provided in the methods for the work to be reproduced?

The fabrication of the device architecture and measurements of the UV detectors are discussed thoroughly in the manuscript, making the reproduction of similar deep UV photodetectors more straightforward.

Detailed technical comments with respect to the manuscript:

I have the following technical comments. If the authors can address these comments while revising the manuscript, the revised manuscript can be considered for publication.

1. Authors are requested to revise the abstract as follows: they should justify the choice of MoS₂, FLG, and hBN in short in the abstract itself. They should also clarify in the abstract why they have explored other materials, like ReS₂, Al₂O₃, Au, Ag, etc, in their work.

2. The authors have said, "By introducing a dielectric barrier layer (e.g., hBN, Al₂O₃) to control the barrier height between the photon absorption layer and charge transport layer, we achieved excellent spectra selectivity for UVB to UVC photodetection, with a tunable cut-off wavelength from 440 to 300 nm," However, hBN also absorbs deep UV (<https://doi.org/10.1002/adom.202100342>, <https://doi.org/10.1038/srep42973>). How can authors rule out the possibility of generating photogenerated charge carriers in the barrier layer? To support the claim, the author should provide the comparative absorption spectra of FLG and hBN.

3. In Figure 2 (e-f), please clarify whether the +V_g and -V_g pulses are applied at the source or the back gate. If the pulses are applied at the back gate, please show them near the back gate in your schematics.

4. The authors should provide the band diagrams corresponding to the configurations of Figure 2 (e)- Figure 2(g). This will enhance the clarity for the readers. Please refer to Figure 2 of the following paper for guidance: doi:

10.1021/acs.nanolett.6b04449

5. Figure 3(f) shows the incoming light incident from the Si side, which may be misleading as the Si/SiO₂ substrate is not transparent enough to pass the incident light and produce photogenerated charge carriers in FLG. This Figure also contradicts Figure 2(g) of the paper. Hence, please revise Figure 3(f).
6. The authors have said, "UV irradiance of solar light with results aligning well with the Meteorological agency's weather report." In this work, the incident light passes through the MoS₂ (Figure 1 (a)), supplementary Fig. 16). But, MoS₂ can absorb the incident visible component of the solar light, generate light-induced carriers, and interfere with the F-N tunneling signal; how could it be possible to primarily detect the deep UV and make the photodetectors insensitive to visible light? The author have also mentioned that the voltage/ potential of the FLG layer under UV irradiation has been monitored by resistance changes in the MoS₂ layer. Won't that be affected by visible light?
7. "As shown in Fig. 3a, the electron (hole) tunneling occurs when the voltage is larger than the threshold voltage in the dark, corresponding to the FNT under a high electric field." This looks acceptable for Figure 3(a). However, with respect to Figure 3 (b), the authors have also mentioned that electron tunneling occurs with the assistance of photon excitation; what is the measurement configuration, and why is 0 V to -4 V ITN modulated under illumination? Unlike the previous report (doi: 10.1021/acs.nanolett.6b04449), are the photocurrent and dark current the same in the FNT regime?
8. Figure 4 (a) shows the light-intensity-dependent speed of response of the fabricated device. Please explain the origin of this phenomenon.
9. With respect to extended data table 1, why is the rise time of the detector not superior to that reported elsewhere? As the supplementary Figure 6 (b) shows, this device has minimum charge traps at the interface. Then why does the rise time not reflect this?
10. In supplementary Figure 18, the authors have mentioned the presence of a "UV filter applied to avoid the impact of the visible light from the voltaic arc". However, wide-band-gap-semiconductor-based UV detectors, as reported in references 10-26, do not require such arrangements (<https://doi.org/10.1038/s41578-023-00583-9>). Please clarify.
11. The author should also discuss how their manuscript differs from other similar works, e.g, doi: 10.1021/acs.nanolett.6b04449, 17, 453–459 and doi: 10.1109/LPT.2020.2993444.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors revised the manuscript according to the reviewer's suggestions.

Reviewer #2

(Remarks to the Author)

Although we appreciate your efforts in addressing the reviewers' concerns and proposing a novel design for the photodetector, there are still several critical issues that need to be addressed before the paper can be considered for publication. Below, I summarize the main points that need further clarification:

1. While your work introduces a complex structural design and a novel working principle, the explanation of the underlying mechanism remains incomplete and lacks sufficient evidence to convince the readers. Additionally, the limited change in the material work function and the barrier concentration around 3.0 eV do not provide generalizability, which needs to be further discussed.
2. Figure 3b show a tunneling current in the pA range, while the channel gain current reaches the μ A level (Figure 3g). However, the source of the gain current remains unclear. We request a detailed explanation of how the gain current is generated in the device.
3. It is unclear why previous experimental data were removed. Was the data deemed unreliable? The update of the packaging method only shows a limited improvement in the device's response time. Your paper presents a large amount of data indicating that the device response time is in the second range, which is slower than common detectors by almost three orders of magnitude. Please provide an explanation for why packaging can enhance the response speed and whether the artificially set compensation current value is appropriate for achieving faster response.
4. There are labeling errors in Figure S18. Moreover, the device noise is close to the lowest testing limit of the equipment, and it is necessary to provide data for the system's base noise. For the detectivity results, it is essential to include the effective area of the device.
5. It is important to present the UV-visible absorption spectra for materials like graphene and MoS₂ used as absorption layers, as this would provide further insights into the device's performance.
6. Generally, the generation of photogenerated carriers is not directional, the mechanism driving the effective separation of photogenerated carriers and the spontaneous tunneling of individual carriers into the channel layer remains unclear. A

detailed explanation of this process is crucial for understanding the functioning of the device.

7. According to the data presented in Figure 4c, the device's responsivity linearly changes with incident light intensity over a wide range. However, you suggest that interface defects have no impact on the device performance, which contradicts the data showing significant defects. Stronger evidence and explanation are required to clarify this discrepancy.

8. In Figure R3/R5, if the absorption layer does not respond to light in the 254-365 nm wavelength range, then it seems that the absorption layer does not contribute to the photoelectric response. Furthermore, the photogenerated current does not appear to come from the absorption layer or the channel layer. A clear explanation of where the photocurrent originates is necessary.

9. Due to these significant unresolved issues, we cannot recommend your manuscript for publication at this time. We encourage you to address these concerns in a more comprehensive manner and re-submit your work after further revisions.

Reviewer #3

(Remarks to the Author)

Concerns regarding the manuscript- the hBN-related UV absorption, the mismatch between the text and the schematic, the band diagram for enhanced clarity, and the MoS₂-related visible-range absorption in the photodetectors have been suitably addressed.

Version 2:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

The revised paper can be accepted.

Open Access This Peer Review File is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made.

In cases where reviewers are anonymous, credit should be given to 'Anonymous Referee' and the source.

The images or other third party material in this Peer Review File are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

To view a copy of this license, visit <https://creativecommons.org/licenses/by/4.0/>

Response to the reviewers' comments

We sincerely thank all three Reviewers for their thorough review and positive feedback on the manuscript. We are pleased that they consider our work to be of high quality and interest. The reviewers have raised important points regarding the manuscript and below we present our point-by-point responses to reviewers' queries. We our revisions and clarities in the revised manuscript, we hope the manuscript is now suitable for publication in Nature Communications.

Response to Reviewer #1:

1. The authors investigated new photodetection scheme based on tunneling barrier modulation rather than the band gap of the absorber material, and achieved highly sensitive DUV photodetection using the two-dimensional van der Waals heterostructures. This is interesting research.

→ We thank for the reviewer's carefully reviewing our manuscript and found the research interesting. As the reviewer point out, we have demonstrated a new photodetection scheme based on the tunneling-barrier controlled photodetector, it shows highly sensitivity for the DUV light. It offers a versatile and scalable approach for UV photodetection across a wide range of materials, showing great potential for future applications.

2. How does the performance change with varying hBN thicknesses?

→ Thank the reviewer for the question. We have fabricated devices with varying thickness, and finally chose the hBN with thickness of approximately 13 nm as a good candidate. Here we show the result of devices with three different thicknesses. First, the MoS₂/hBN/FLG device made with a 9-nm-hBN shows a high resistance switching ratio of 10^7 , the dark current at high resistance state (HRS) is as low as 10^{-13} A, which is one magnitude lower than the that made of 13 nm-thick hBN, as shown in Fig. R1a. However, the high resistance state of 9-nm-hBN is not easily to be stabilized as shown in Fig. R1b. Second, the device made of 20-nm-thick hBN shows a reduced resistance switching ratio when increases the thickness of hBN, therefore the dark current at the HRS is enhanced, which is not beneficial for the photodetection. Therefore, we chose the thickness of 13-nm-hBN which shows a relatively low dark current at HRS and also could be stable for a long time (>10000 s). To make it clear for the readers, we have added the Fig. R1 to the Supplementary information Fig. 2.

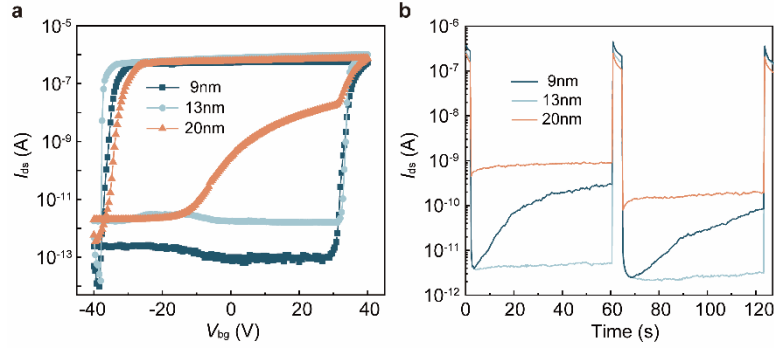


Fig. R1 characterization of MoS₂/hBN/FLG devices made with different thicknesses of hBN. (a) Transfer curves of the devices with different thicknesses of hBN layer, 9 nm, 13 nm and 20 nm. (b) The HRS stability test in the dark for devices with different thicknesses hBN layer.

3. How does defects at the interface of MoS₂/hBN/FLG affect the photodetector performance?

→ The defects usually have an impact on the electric characteristics of 2D materials. In this work, we tried our best to ensure the high quality of the heterostructure devices by using the dry transferring technique and removing the residue pastes by annealing the samples at 300 °C in N₂ atmosphere after transferring a new layer. Moreover, the highly DUV photoresponse of the device have been proved not to be related with the interfacial defects through the comparison experiments. We compared the photoresponse of the device when the backgate voltage is applied on silicon substrate and that of the device when backgate is applied on FLG layer (Fig. S7)), and that of the device when the FLG layer is grounded under the same light condition (Fig. S8a). It is clear that the photoresponse is disappeared for the last two cases, even though interface has not been changed. This result shows the UV photoresponse layer is due to the charges stored at the FLG but not due to the interface of the device.

Response to Reviewer #2:

Li et al report an interesting result on tunneling barrier controlled ultrasensitive deep ultraviolet photodetector based on van der Waals heterostructures, demonstrating application potential for solar-blind detection application. The authors have developed a complex device structure and operating process that enhances the optical gain of traditional devices, however, the reported response time of 0.5 s is significantly longer than what is typically expected for tunneling-type electronic devices, which are known for their high-speed response. This long response time undermines the advantages of the proposed device and is difficult to accept. several critical concerns lead me to recommend rejection of the manuscript:

→ We thank the reviewer for her/his careful reading of our manuscript and for finding our results interesting with significant potential for applications. However, we believe there

may have been some misunderstanding regarding the working mechanism of our device. In response, we have carefully revised and improved the manuscript to address these points.

Specifically, we have enhanced the response time of our device through two approaches. By setting an appropriate compliance current, the response time can reach ~60 ms for 254 nm UV light with an intensity of 120 $\mu\text{W}/\text{cm}^2$, without compromising its sensitivity to weak light. Additionally, by packaging the device using a commercial TO-39 with a lens cap, the response speed can be further improved by a factor of 7. As a result, the response speed of our photodetector is now comparable to that of many UV photodetectors made with ultrawide bandgap semiconductors, as shown in Extended Data Table 1. This performance meets the requirements for various applications, such as serving as a UV power meter.

We understand that the reviewer may have expected the device to exhibit ultrafast response speeds due to the electron-tunneling. However, in our device, we do not directly measure the tunneling current as the photocurrent. Instead, to achieve a high responsivity, the MoS₂ channel current is regarded as the photocurrent. The MoS₂ channel current saturates only after the potential of the FLG layer recovers to zero, following the complete depletion of stored electrons. Stronger light depletes the stored charges more quickly, resulting in a faster response speed. In the revised manuscript, we demonstrate that the response time could reach 60 ms under relatively weak light conditions (120 $\mu\text{W}/\text{cm}^2$). This response time could be significantly faster for stronger UV light, as it is inversely proportional to the power density of incident light.

A detailed point-point response has been provided to address the reviewer's questions. We hope the revised manuscript clarified these issues and resolve the reviewer's concerns.

1. The two-dimensional materials generally pose challenges for ultraviolet light absorption and photoelectric conversion efficiency. Therefore, using two-dimensional materials as the light absorption layer does not seem optimal. Although the cutoff response wavelength in tunneling devices is determined solely by the barrier height of the heterojunction (independent of material bandgap), selecting wide bandgap semiconductors with high absorption coefficients for the absorption layer would better facilitate efficient photoelectron generation and improve the optical gain of UV detectors.

→ We sincerely thank the reviewer for the valuable comments. In this work, we demonstrated a novel approach to realize the UV selective detection through barrier controlled heterostructures. This mechanism enables the use of general bandgap semiconductors for DUV photodetection, thereby offering broader material selection flexibility. We agree that selecting wide bandgap semiconductors with high absorption coefficients as the absorption layer could potentially enhance optical gain. However, these wide bandgap semiconductors often face challenges related to the defect formation during growth process, which can adversely impact their performance. The heterostructure-based approach we propose seeks to address these limitations by leveraging high-quality interfaces and material compatibility. If high-quality wide bandgap semiconductor thin films become readily available, combining them with our

heterostructure design would indeed be a promising direction for further improving UV photodetection performance.

2. The design where the incident light enters from the top while the photosensitive absorption layer is positioned beneath the transport layer is unusual for photodetector structures. An explanation for this design choice is necessary.

→ We appreciate the reviewer's insightful comment. Indeed, the design where the FLG layer is positioned beneath the transport layer may appear unconventional. To address this, we also tested a device structure where the FLG layer is on top, forming an FLG/hBN/MoS₂ heterostructure on the SiO₂/Si substrate. In the FLG-on-top structure, the equivalent capacitance between MoS₂ and Si channel is shown Fig. R2b. Since $C_{\text{MoS}_2/\text{Si}}$ is smaller than $C_{\text{Si/FLG}}$ and $C_{\text{FLG/MoS}_2}$, so the carrier density in MoS₂ layer is controlled by the double-capacitance coupling between the $C_{\text{Si/FLG}}$ and $C_{\text{FLG/MoS}_2}$, which is same with the FLG layer in between.

$$C_{tot} = \frac{C_{\text{Si/FLG}} \cdot C_{\text{FLG/MoS}_2}}{C_{\text{Si/FLG}} + C_{\text{FLG/MoS}_2}}$$

Therefore, the device shows similar transfer characteristic and photoresponse behavior with that FLG is in between.

Since the few-layer TMDCs and hBN used in our design exhibit high transparency to UV light, enabling highly efficient photoresponse even with the absorption layer beneath the transport layer. In the manuscript, we chose to present the FLG-in-between structure to emphasize the carrier migration mechanism between different layers while avoiding the complexity of explaining double-capacitance coupling.

As our results demonstrate, the photoresponse performance remains robust regardless of the layer configuration.

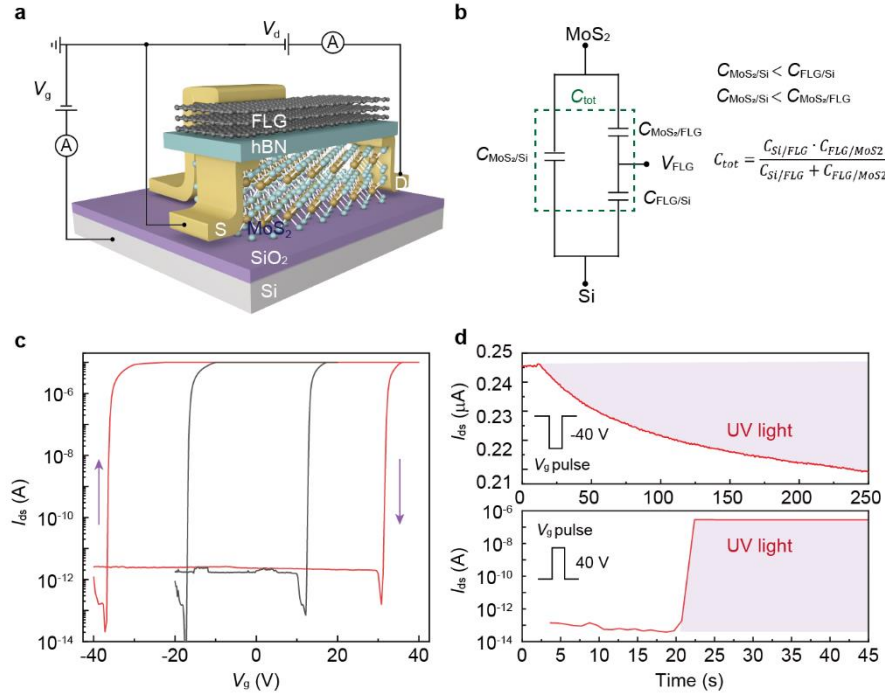


Fig. R2. The FLG/hBN/MoS₂ structure on the SiO₂/Si substrate. (a) Schematic diagram of FLG/hBN/MoS₂ device. (b) The equivalent capacitance circuit between the MoS₂ and Si layer. The total capacitance is determined by the capacitance between MoS₂ and FLG ($C_{\text{MoS}_2/\text{FLG}}$) and capacitance between FLG and Si ($C_{\text{FLG}/\text{Si}}$). (c) Transfer curves under different V_g sweep ranges. The hysteresis window is increase with the sweep V_g range. (d) The PPR and NPR photoresponse for the FLG/hBN/MoS₂ device on 405 nm.

3. Without tunneling involvement, it remains unclear how to differentiate between the photogenerated current in the transport layer and the contribution from tunneling to the final photocurrent gain.

→ Thanks for the question. We measured the intrinsic photoresponse of the MoS₂ layer when the FLG layer is grounded (as shown in Supplementary Fig. 7). Under the same UV light condition, the MoS₂ exhibited near zero photoresponse, indicating that the photogenerated current in the transport layer is ignorable. The final photocurrent is regulated by the potential of the FLG layer. This potential is influenced by the total number of electrons migrating from the FLG layer to the MoS₂ layer under UV irradiation over time. Furthermore, the observed non-volatile photoresponse confirms that the channel current is governed by the potential of FLG layer rather than the photogenerated current.

4. In line 162 of the manuscript, please provide specific values for the uncommon parameters $\frac{\mu_{\text{FLG}}}{\mu_{\text{Si}}}$ and $\frac{\mu_{\text{FLG}}}{\mu_{\text{Si}}}$ used in the equation.

→ We appreciate the reviewer's feedback regarding the parameters mentioned in line 162 of the manuscript. However, it seems there may be a formatting or typographical issue in

the comment. Based on the context, we are uncertain if the reviewer is referring to the effective masses of electrons and holes in hBN.

In this work, we consistently use the same values of effective mass of electrons and holes in hBN for all calculations. According to the references (“*Physica B Condens* **585**, 412125 (2020)” and “*Adv. Sci.* **8**, 2101572 (2021)”), the effective mass of electrons and holes in the hBN are estimated to be 0.064 and 0.25, respectively. Using these values, we are able to achieve consistent results in our manuscript across different devices, such as MoS₂/hBN/FLG, ReS₂/hBN/FLG, MoS₂/hBN/Au, as shown in Fig. 3g and 3h.

We believe these values provide a reasonable and reliable estimate for the effective masses in hBN. We have specified these values in the revised manuscript for clarity.

5. Is a gate voltage necessary to drive the photogenerated electrons/holes from the absorption layer to the transport layer? In Figure 2g, it is stated that under UV illumination, photogenerated electrons in the absorption layer tunnel into the transport layer, resulting in an extremely high photocurrent. However, without gate voltage assisting the tunneling process, clarification is needed regarding the driving force for the electrons to cross the barrier and inject into the transport layer.

→ In the working mechanism of the photodetector, the photogenerated electrons and holes of the absorber layer do not require the gate-voltage to move into the transport layer. However, the application of a gate-voltage is crucial for resetting the channel into HRS. The gate-voltage pulse enables the injection of electrons into the absorbing layer, creating a negative potential in FLG layer to help maintain a low dark current in the detector.

Under UV illumination, the photogenerated electrons and holes in the absorption layer absorb the energy of the incoming photons, which excites them to higher energy states. This process significantly enhances the probability of carrier tunneling through the dielectric layer (such as hBN) into the transport layer, thereby boosting the optical response of the detector. In this process, the photon energy is the primary driving force that facilitates the migration of the carriers, while the gate voltage ensures the device operates with minimal dark current.

We hope this clarifies the role of the gate voltage and the mechanism of carrier migration in our photodetector.

6. Supporting Data for Figure S7: Providing time-resolved photoresponse data for VFLG at 0 V and 3V conditions would add more persuasive evidence. Additionally, does VFLG>0 favor obtaining a larger optical gain?

→ We appreciate the reviewer's suggestion to provide time-resolved photoresponse data for V_{FLG} at 0V and 3 V. To address this, we have conducted the requested measurements, as shown in **Fig. R3**. The results shown that an increased gate-voltage ($V_{\text{FLG}} = 3\text{V}$) enhances the dark current, as indicated by the transfer curve. Additionally, $V_{\text{FLG}} = 3\text{V}$ does not lead to a higher photogain; instead, it primarily increases the dark current. More

importantly, under both V_{FLG} at 0V and 3 V, the device does not exhibit significant photoresponsivity to UV light.

These results further confirm that the high photoresponse observed in our device is not due to the intrinsic photoelectric properties of the MoS₂ layer or the hBN layer. We have included these findings in the revised manuscript and supporting data to strengthen the evidence for our proposed mechanism.

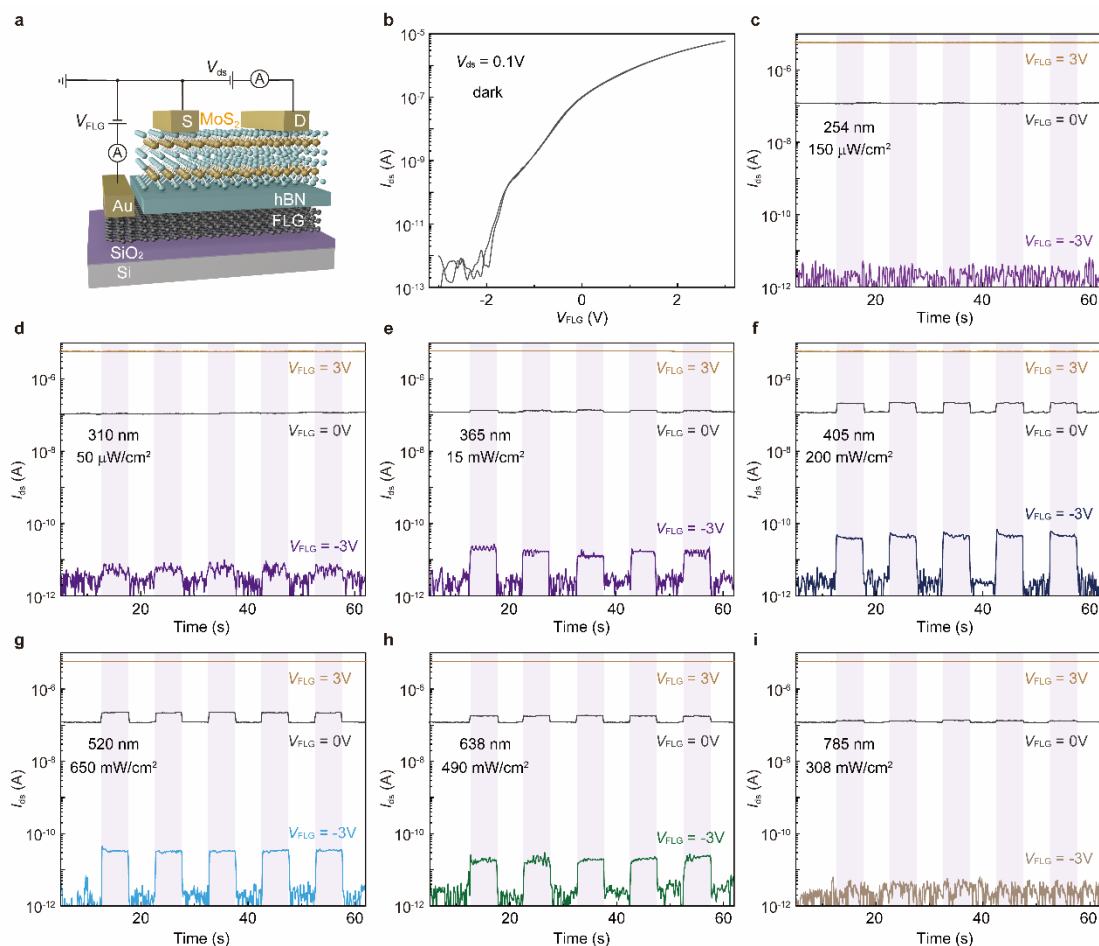


Fig. R3. Photoresponse of the device when V_{FLG} is applied on FLG layer (a) The measurement configuration when the gate voltage is applied through the FLG layer (b) Transfer curves when the gate voltage is applied through the FLG layer. (c-i) Time-resolved photoresponse of MoS₂ channel when $V_{\text{FLG}} = -3$ V, 0 V and 3 V for different wavelength light, (c) 254 nm, (d) 310 nm, (e) 365 nm, (f) 405 nm, (g) 520 nm, (h) 638 nm, (i) 785 nm light.

7. The manuscript needs thorough language proofreading to ensure accuracy and fluency in academic writing, as well as adherence to the journal's formatting guidelines.

→ We appreciate the reviewer's suggestion to improve the language and formatting of the manuscript. We have carefully proofread the text to enhance its accuracy and fluency in academic writing. Additionally, we have revised the manuscript to fully adhere to the journal's formatting guidelines.

8. Regarding Fig. 15 in the supporting material, the current at $V_d = 0.1$ V is only an order of

magnitude lower than at 0 V with frequency more than 1000 Hz, which is quite unusual. Moreover, comparing it with the dark current data in Fig. 2h of the main text, the current density may have been underestimated.

→ Thanks for pointing it out. To address this, we have re-examined the noise spectrum results and conducted additional measurements on several samples.

Our finding indicates the noise level is below 10^{-26} (A^2/Hz) across different bias voltages ranging from 0.001 to 1V (**Fig. R4**). Furthermore, the noise level remains relatively unchanged with varying applied bias voltages. For comparison, we also measured the noise spectrum of a single MoS₂ device, which exhibits a relatively higher noise level that increases with applied bias. The MoS₂/hBN/FLG device demonstrates significantly reduced noise, attributed to the negative potential (~ -5 V) in the FLG layer when the device is in a high-resistance state. This negative potential strongly suppresses both the dark current and the noise level.

Regarding the dark current, our measured value is approximately 10^{-11} A with typical bias voltages of 0.1~0.5 V. Using this dark current, the estimated noise level, calculated as $S = \sqrt{2qI_{\text{dark}}}$, is approximately $\sim 10^{-15} \text{ A} \cdot \text{Hz}^{-1/2}$. This estimated noise level is much lower than the noise spectra measured through the noise analyzer. Thus, there is no underestimation of the noise level for our device.

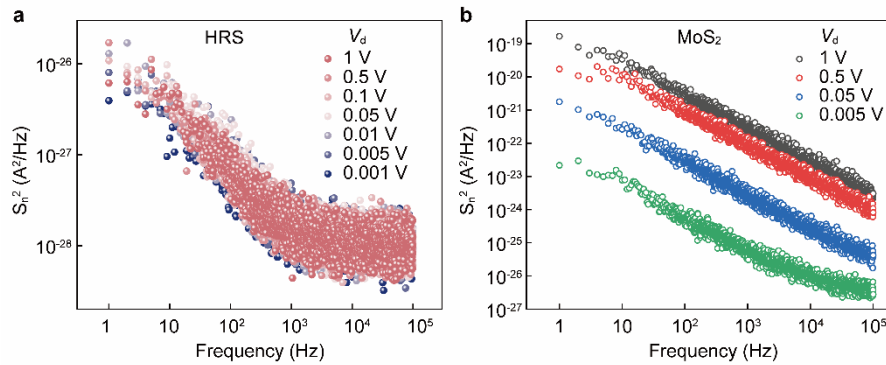


Fig. R4. Noise spectra of the MoS₂/hBN/FLG (a) and MoS₂ (b) device at varying applied biases. The MoS₂/hBN/FLG device is regulated into high resistance state.

9. It is unreasonable to simply compare Figure 2g and Figure 7 in the supporting materials to demonstrate the advantages of the ultra-high photoresponse of tunneling devices. They do not operate in similar modes and lack reasonable controlled variables. Figure 2g is a floating gate device, while Figure 7 in the supporting materials operates similarly to a transistor. The photocurrent generated by the absorption layers, such as graphene, seems unlikely to produce such a large photocurrent as a source of tunneling current; please provide the response curves of graphene and other light-absorbing materials under violet light.

→ Thank you for your detailed comment. The comparison between Fig. 2g and Fig. S7 demonstrates that the strong photoresponse in the UV region is not due to the MoS₂ material but is instead attributed to the excitation of stored charges in the FLG layer. When UV photoexcitation alters the potential of the FLG layer by facilitating electron tunneling,

it modulates the conducting states of the MoS₂ layer. The photocurrent, measured as the channel current, is tuned through capacitive coupling between the FLG and MoS₂ layers.

Additionally, we investigated the UV photoresponse of hBN and FLG phototransistors. The FLG device shows negligible photoresponse due to its high dark current. Similarly, the hBN material exhibited only a minimal photoresponse in the UVC region and no detectable response to light with wavelengths longer than 254 nm. These results conclusively rule out significant contributions to the photoresponse from the FLG and hBN materials.

This revision maintains the technical content while streamlining the explanation for better readability.

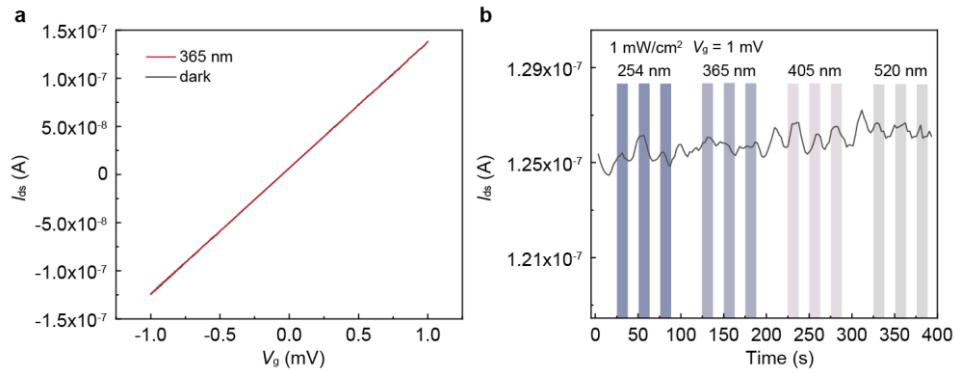


Fig. R5. Photoreponse characteristics of FLG device on SiO₂/Si. (a) I-V output curve; (b) I-T data, measured under a light intensity of 1 mW/cm² with wavelengths of 254 nm, 365 nm, 405 nm and 520 nm. None exhibited a significant photoresponse.

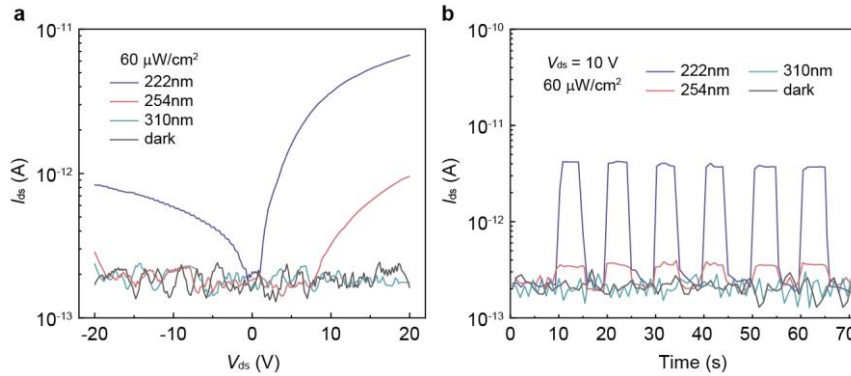


Fig. R6. Photoreponse characteristics of BN device on SiO₂/Si. (a) I-V output curve; (b) I-T data, measured under a light intensity of 60 μ W/cm² with wavelengths of 222 nm, 254 nm, and 310 nm. Notably, the conditions at 310 nm did not produce a significant photoresponse.

10. In summary, the operational mode of this structure is more akin to a memory that is electrically written and optically erased. This structure is better suited to utilize the principles of optical floating gate transistors for memory instead of detectors.

→ Definitely, the device can be used as a memory device. However, the high responsivity of the device also makes it an outstanding UV photodetector. It can detect 254 nm UVC

light at intensity as low as $0.1 \mu\text{W}/\text{cm}^2$, outperforming commercial thermal-electric power meters. Furthermore, we have demonstrated its potential application as a UV power meter. Its full compatibility with CMOS processes also enables great opportunities for large-scale production.

Additionally, we have demonstrated a barrier-engineered approach to achieve DUV photodetection with this device, which could be extended to other material systems. This demonstrates a promising pathway to develop more high-performance UV photodetectors. With optimized design, the floating gate may not even be necessary for such devices in the future.

Response to Reviewer #3

-What are the noteworthy results?

The authors have presented deep ultraviolet (DUV) photodetection with impressive performance metrics (e.g., responsivity $\sim 4 \times 10^4$, $I_{\text{on}}/I_{\text{off}} \sim 10^6$) using MoS₂ as the charge transport layer and few-layered graphene (FLG) as the photon absorption layer, separated by a hexagonal boron nitride barrier layer. The authors claim that the novelty lies in the DUV photodetector's cut-off wavelength tuneability through an electrically modulated electron F-N tunnelling barrier, which is not limited to the bandgap of the absorption material. Please refer to the comments below.

→ Thanks for the reviewer's careful reviewing our work and think it shows an impressive performance. As the reviewer pointed out, we have demonstrated a new photodetection scheme which could realize high performance UV photodetection without relying on the wide bandgap semiconductors. It provides a novel approach to overcome the challenges encountered in UV photodetectors based on wide/ultrawide semiconductors, such as material growth and effective doping.

- Will the work be of significance to the field and related fields?

Yes, because the author has presented results on deep UV detectors with outstanding performance metrics.

→ Thanks for the positive evaluation of the significance of our work.

-How does it compare to the established literature? If the work is not original, please provide relevant references.

The work is original. However, the device architecture presented in this work has similarities with the other reports (DOIs mentioned below): doi: 10.1021/acs.nanolett.6b04449, 17, 453–459, doi: 10.1109/LPT.2020.2993444

→ As the reviewer pointed out this work is original. And we have clearly discussed the differences between our work with the mentioned references in the response to question 11. Our device works in a different mechanism from that of the referred works, and has shown its universality and can be widely applied.

- Does the work support the conclusions and claims, or is additional evidence needed?

The author has supported their claims with data; however, additional evidence is needed.

Please refer to my other comments to know more about the required revisions.

→ Thanks for the reviewer's positive evaluation. We have added more experimental data as requested by the reviewer, we believe these results could eliminate the reviewer's concern.

- Are there any flaws in the data analysis, interpretation, and conclusions? - Do these prohibit publication or require revision?

To support their claim, the author should revise the manuscripts with some additional evidence, such as hBN's (barrier layer) deep UV absorption, charge carrier generation, etc. Please refer to my other comments below to learn more about the required revisions.

→ We have revised the manuscript according to the reviewer's suggestion. We believe it will be clearer to the readers. The detailed revision has been listed in the summary of changes in the end of the response letter.

- Is the methodology sound? Does the work meet the expected standards in your field?

Yes, the methodology is sound, and the author has supported the claim with relevant data; however, in a few areas, it still needs clarity before publication. Please refer to my other comments below to know more about the required revisions.

→ Thanks for the reviewer's positive evaluation. We have revised the manuscript to make it much clearer to the general reader.

- Is there enough detail provided in the methods for the work to be reproduced?

The fabrication of the device architecture and measurements of the UV detectors are discussed thoroughly in the manuscript, making the reproduction of similar deep UV photodetectors more straightforward.

→ Thanks for the reviewer's positive evaluation.

Detailed technical comments with respect to the manuscript:

I have the following technical comments. If the authors can address these comments while revising the manuscript, the revised manuscript can be considered for publication.

→ Thanks for the reviewer's positive evaluation. We believe our revision of the manuscript could remove the reviewer's concerns, and improved the manuscript, therefore it could meet the demands of the journal.

1. Authors are requested to revise the abstract as follows: they should justify the choice of MoS₂, FLG, and hBN in short in the abstract itself. They should also clarify in the abstract why they have explored other materials, like ReS₂, Al₂O₃, Au, Ag, etc, in their work.

→ Firstly, the emerging two-dimensional van der Waals layered materials shows excellent

optoelectrical properties, particularly, they can be stacked into heterostructures with high interfaces quality without lattice mismatch. Thus, it becomes a very good system to verify new heterostructure devices, while the traditional heterostructure growth requires many efforts on the material growth and considering the lattice matching between different layers. Therefore, in this work we selected the 2D materials as a platform to validate the barrier controlled DUV photodetection mechanism.

The few-layered MoS₂ and ReS₂ was chosen as the charge transporting layer due to their high carrier mobility and tunable low dark current, while the FLG was chose as the charge storage and photon absorption layer due to its broad spectrum photoresponse. And hBN is selected as the barrier layer due to its excellent dielectric properties. After showing the high performance of the device, we further extended the photodetection scheme to a wide range of materials, utilizing different charge transporting layer (e.g., MoS₂, ReS₂), barrier layer (e.g., hBN, Al₂O₃), and photon absorption materials (e.g., FLG, PdSe₂, Au, Pd), showcasing its broad adaptability and potential for extensive application.

We have added related explanation in the revised manuscript.

2. The authors have said, “By introducing a dielectric barrier layer (e.g., hBN, Al₂O₃) to control the barrier height between the photon absorption layer and charge transport layer, we achieved excellent spectra selectivity for UVB to UVC photodetection, with a tunable cut-off wavelength from 440 to 300 nm,” However, hBN also absorbs deep UV (<https://doi.org/10.1002/adom.202100342>, <https://doi.org/10.1038/srep42973>). How can authors rule out the possibility of generating photogenerated charge carriers in the barrier layer? To support the claim, the author should provide the comparative absorption spectra of FLG and hBN.

→ The hBN barrier layer is an ultrawide bandgap material with a bandgap of ~6 eV. Therefore, it shows intrinsic photoelectric effect for deep ultraviolet with very short wavelength (intrinsic photo absorption occurs for wavelengths shorter than ~206 nm). We have tested the photoresponse of the hBN material directly, it only shows weak photoresponse for wavelengths shorter than 254 nm. The photocurrent for 60 μW/cm² 222 nm UV light is approximately 4×10⁻¹²A, as shown in **Fig. R4**. It is similar to the results reported in the *Adv. Optical Mater.* **9**, 2100342 (2021). The intrinsic photoresponse of hBN is very low comparing to UV photoresponse of our heterostructure device, and the spectral response is also completely different from our device. Therefore, the high photoresponse of our device is not attributed to the hBN barrier layer.

3. In Figure 2 (e-f), please clarify whether the +V_g and -V_g pulses are applied at the source or the back gate. If the pulses are applied at the back gate, please show them near the back gate in your schematics.

→ In Fig. 2 (e-f), the +V_g and -V_g pulses voltage are applied to the back gate, as it is named as gate-voltage. To eliminate the confusion, we have replotted the Fig. 2e and 2f. The pulse voltage symbol has been displayed near the back gate.

4. The authors should provide the band diagrams corresponding to the configurations of Figure 2 (e)- Figure 2(g). This will enhance the clarity for the readers. Please refer to Figure 2 of the following paper for guidance: doi: 10.1021/acs.nanolett.6b04449

→ Thanks for the suggestion, we have added the band diagrams corresponding to the moment of applying positive gate-voltage pulse and negative gate-voltage pulse in Fig. 2e and 2f. The electrons and holes are trapped in the FLG layer with the help of gate-voltage pulses. In Fig. 2g, we have also plotted the electron's and hole's tunneling out the FLG under the UV irradiation. Hopefully, this will be clear for the readers to understand the three different processes under gate-voltage pulse and light illumination.

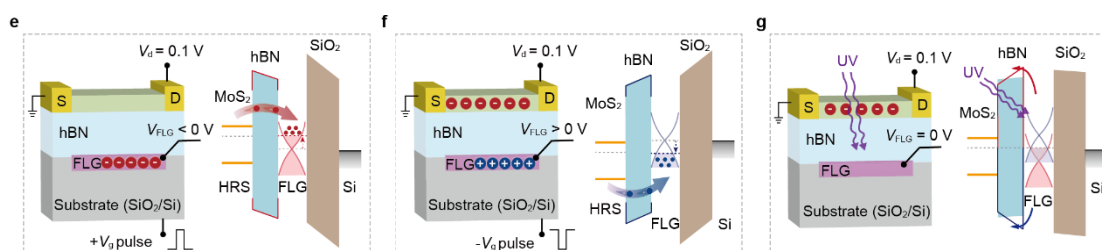


Fig. R7. Revised Fig. 3e to 3g, with the corresponding band diagram added.

5. Figure 3(f) shows the incoming light incident from the Si side, which may be misleading as the Si/SiO2 substrate is not transparent enough to pass the incident light and produce photogenerated charge carriers in FLG. This Figure also contradicts Figure 2(g) of the paper. Hence, please revise Figure 3(f).

→ Thanks for the suggestion. We have revised the Fig. 3f to eliminate the confusion about the light incident direction in the revised manuscript.

6. The authors have said, “UV irradiance of solar light with results aligning well with the Meteorological agency’s weather report.” In this work, the incident light passes through the MoS2 (Figure 1 (a)), supplementary Fig. 16). But, MoS2 can absorb the incident visible component of the solar light, generate light-induced carriers, and interfere with the F-N tunneling signal; how could it be possible to primarily detect the deep UV and make the photodetectors insensitive to visible light? The author have also mentioned that the voltage/potential of the FLG layer under UV irradiation has been monitored by resistance changes in the MoS2 layer. Won’t that be affected by visible light?

→ Thank you for your question. Based on the spectral response of the MoS2/hBN/FLG device, the cutoff wavelength of the device is approximately 440 nm. To measure UV irradiance from the sunlight, a UV filter was used to remove the impact of visible light as it has been mentioned in the main text. The MoS2 exhibits minimal absorption of UV light, and its influence on the UV photodetection is negligible. This has been confirmed through a series measurement of the photoresponse (Fig. S7, Fig. S8, and Fig. S9).

Importantly, we obtained the photoresponse time verse power intensity relationship for the device with the same configuration (MoS2 layer on the top), thus the influence of

MoS₂'s absorption can be excluded in calibrating the UV intensity from the sunlight. In this configuration, the MoS₂ channel current is mainly controlled by the potential change in FLG layer via capacitance coupling, with the intrinsic photoresponse of MoS₂ layer being negligible in the UV region. Additionally, under weak light illumination, the device demonstrates high rejection ratio (R_{250}/R_{450}) of up to 10^6 for wavelengths beyond 440 nm.

7. "As shown in Fig. 3a, the electron (hole) tunneling occurs when the voltage is larger than the threshold voltage in the dark, corresponding to the FNT under a high electric field." This looks acceptable for Figure 3(a). However, with respect to Figure 3 (b), the authors have also mentioned that electron tunneling occurs with the assistance of photon excitation; what is the measurement configuration, and why is 0 V to -4 V ITN modulated under illumination? Unlike the previous report (doi: 10.1021/acs.nanolett.6b04449), are the photocurrent and dark current the same in the FNT regime?

→ Fig. 3b and Fig. 3a were measured under the same condition, with Fig. 3b was measured under light illumination. When $|V_g|$ is smaller than threshold voltage (5V), the tunneling current is almost negligible because FN tunneling and directly tunneling are unlikely to occur in the dark. Under these conditions, the photo illumination assists electrons tunneling leading to the increase in I_{TN} . However, when V_g is larger than the threshold voltage for FN tunneling, the dark current increases rapidly with the voltage. In this region, photon assisted electron-tunneling contributes only a small fraction to the total tunneling current, making it difficult to observe a clear distinction between the dark current and the photocurrent. To address this, we have provided a linear-scale plot of I_{TN} under dark and illuminated conditions, where a substantial difference can still be observed.

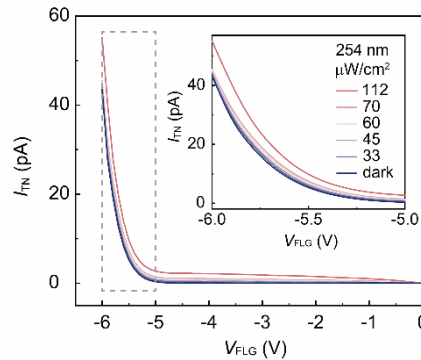


Fig. R8. The linear scale plot of the I_{TN} under 254 nm light illumination with varying intensities.

8. Figure 4 (a) shows the light-intensity-dependent speed of response of the fabricated device. Please explain the origin of this phenomenon.

→ Since the photocurrent of the MoS₂ layer saturates until the charges stored in the FLG layer are fully excited out, i.e. the potential of the FLG layer reaches zero, as shown in Fig. 2e. In this process, the rising time (t) of the photocurrent is inversely proportional to the light intensity (P), because higher intensity leads to a faster depletion of the stored charges

in FLG layer. And due to the number of charges stored in the FLG layer are constant, the product ($P \cdot t$) should be a constant as well (**Fig. 4a**). This is the relationship we used to calibrate the power intensity of incident light.

9. With respect to extended data table 1, why is the rise time of the detector not superior to that reported elsewhere? As the supplementary Figure 6 (b) shows, this device has minimum charge traps at the interface. Then why does the rise time not reflect this?

→ As shown in the updated extended data figure x in the revised manuscript, we achieve a fast response time of 70 ms for 254 nm UV light at an intensity of $300 \mu\text{Wcm}^{-2}$. This response speed is comparable to that of many wide bandgap semiconductors, as detailed in Extended data table 1. Such a response speed meets the requirement for various UV photodetection applications, including power metering.

Moreover, the response time presented in extended data table 1 corresponds to the time at which the photocurrent reaches saturation. This is determined by the time required for carrier depletion in the FLG layer. It is because we are measuring the MoS₂ channel current as the photocurrent rather than directly measuring the tunneling current. The MoS₂ channel current is determined by the capacitance coupling between the FLG and MoS₂, so the saturation occurs only when the charges stored in the FLG layer are fully depleted. This is not indicative of charge trapping at the interface.

Additionally, as shown in the revised Fig. 4b, we can adjust the compliance current during operation. By setting a lower compliance current, the response time can be reduced to 60 ms for 254nm UV light with an intensity of $120 \mu\text{Wcm}^{-2}$ without compromising the device's ability to detect weak light.

10. In supplementary Figure 18, the authors have mentioned the presence of a “UV filter applied to avoid the impact of the visible light from the voltaic arc”. However, wide-band-gap-semiconductor-based UV detectors, as reported in references 10-26, do not require such arrangements (<https://doi.org/10.1038/s41578-023-00583-9>). Please clarify.

→ In the manuscript, we demonstrated UV light measurement from a voltaic arc using the MoS₂/hBN/FLG device. As shown in the **Fig. 3d**, the cut-off wavelength of MoS₂/hBN/FLG device is approximately 440 nm. To fully eliminate the impact of visible light during measurement, we used a UV filter, because the device is still sensitive to short range of visible light (400-440 nm). It is plausible to detect the UV light without a filter if using a device with a cut-off wavelength in the UV region.

As discussed in the introduction, traditional UV selective photodetector often rely on wide- or ultrawide- bandgap semiconductors, which can function without a UV filter. However, these devices face related to material growth, effective doping, and high production costs. This is the motivation behind our proposed approach, which offers a potential more practical and scalable solution for UV photodetection without relying on wide-bandgap materials.

11. The author should also discuss how their manuscript differs from other similar works, e.g, doi: 10.1021/acs.nanolett.6b04449, 17, 453–459 and doi: 10.1109/LPT.2020.2993444.

→ While the structure of our device bears some similarity to the referred works. Our approach differs significantly in several key aspects:

(1) The referenced devices are two-terminal devices that rely on the tunneling current between two semiconducting layers, which necessitates continuous bias voltages for operation. And large bias voltage (e.g. 4V) is required to ensure high photocurrent. In contrast, our device utilizes a three-terminal configuration, where the photocurrent is measured as the channel current, rather than directly measuring the tunneling current. We operate with a very low bias voltage (0.1 V) to detect the photoresponse, while periodic ultrashort gate voltage pulses reset the channel current to a high-resistance state. This design minimizes power consumption and enhances the stability of the device.

(2) Since we are not directly detecting the tunneling current, we can achieve ultrahigh photocurrent due to the capacitance coupling even for extremely weak irradiations. Our device shows large on/off ratio, and highly sensitivity to 0.1 $\mu\text{W}/\text{cm}^2$ UV light. This performance exceeds that of most UV photodetectors.

(3) our device demonstrates a barrier-controlled spectral photoresponse, which allows for UV photodetection without relying on wide-bandgap semiconductors. This is a significant advantage, as the growth and doping of wide-bandgap materials present substantial challenges. By using general bandgap semiconductors, our method offers a more versatile and scalable approach for UV photodetection across a wide range of materials.

Summary of Changes

1. We have added the band diagram showing the electron (hole) tunneling process corresponding to the positive gate pulse or negative gate pulse in Figs. 2e to 2g. And detailed explanations have been added in the revised manuscript, in line 140-150.
2. We have supplied the parameter values of the effective mass of electron and hole in hBN in the revised manuscript during the calculation of the tunneling barrier, in line 165.
3. We have provided the justification of selecting the metal thin film layer as the photon absorption layer in line 220-225.
4. We have revised the Fig. 4b and shown the response time improvement by setting different compliance current, which could shorten the response time of the device to 60 ms for 254 nm UV light with an intensity of 120 $\mu\text{W}/\text{cm}^2$, in line 255
5. We have discussed the photoresponse of our device is different from that of MoS₂ material in line 285, to eliminate the concern regarding the origination of the photocurrent.
6. We have revised the extended data figure 2, where the device is packaged by the TO-39 with a lens cap. The response time could be improved by 7 times, leading to the response time of 70 ms without setting compliance current.

7. We have updated the response time at varying light intensities in Extended data table 1.
8. We have also added the results of the samples made with different thickness of hBN layer in supplementary Figure 2.
9. We have also revised the manuscript according to the Journal's formatting requirement, and checked the writing carefully.

Response to reviewer comments

Reply to Reviewer #2

Although we appreciate your efforts in addressing the reviewers' concerns and proposing a novel design for the photodetector, there are still several critical issues that need to be addressed before the paper can be considered for publication. Below, I summarize the main points that need further clarification:

→ We thank the reviewer for her/his careful evaluating our manuscript. Here we made a point-to-point response to the reviewer's question. We hope the reviewer's concern could be removed after reading to the response letter.

1. While your work introduces a complex structural design and a novel working principle, the explanation of the underlying mechanism remains incomplete and lacks sufficient evidence to convince the readers. Additionally, the limited change in the material work function and the barrier concentration around 3.0 eV do not provide generalizability, which needs to be further discussed.

→ We appreciate the reviewer's feedback and understand the concern regarding the underlying mechanism of the photodetector. Here, we try to summarize the underlying mechanism of the device in a simple way, hope it would be helpful for understanding.

The working mechanism of the photodetector involves two aspects: photocurrent generation and spectral response.

(1) The photocurrent (i.e. the channel current) is determined by the capacitance coupling of FLG layer, this is evidenced by the time-resolved photocurrent changes together with the potential change of the FLG layer (**Fig. 2i and 2j**). That is when the potential of the FLG layer increased from -5 V to 0 V, the electron concentration in MoS₂ layer increases, leading to the large photocurrent.

(2) The photon illumination could change the potential of the FLG layer by assisting the stored electrons to tunnel out. Due to the existing of the tunneling barrier, the photoresponse shows a spectral selectivity with a cutoff wavelength corresponds to the tunneling barrier height. This is evidenced by the tunneling barrier height measurement and spectral response of a series of devices (**Fig. 3**).

Furthermore, as it has been shown in **Fig. 3g and 3h**, we have verified the potential barrier-controlled photo response mechanism in a series of devices made

with different materials (six different combinations). The barrier height can be tuned from 2.8 eV to 4.0 eV, showing a spectral response tuned from 440 nm to 300 nm. The measured barrier heights do not cluster around 3 eV; therefore, we believe this supports the generalizability of our findings.

2. Figure 3b show a tunneling current in the pA range, while the channel gain current reaches the μA level (Figure 3g). However, the source of the gain current remains unclear. We request a detailed explanation of how the gain current is generated in the device.

→As mentioned before, the photocurrent (i.e., channel current) is controlled by the capacitance coupling effect between the MoS₂ and FLG layer. When the potential of the FLG layer changes from -5 V to 0 V, it could induce the electron accumulation in the channel layer. The carrier density can be estimated through the detailed calculation. **Fig. R1** shows the schematic structure of the device and the calculation procedure to estimate the channel current. It's important to note that the primary effect of electron tunneling (I_{TN}) through the barrier layer is to modify the potential in the FLG layer. This tunneling process does not directly contribute to the measured photocurrent in the described working mechanism.

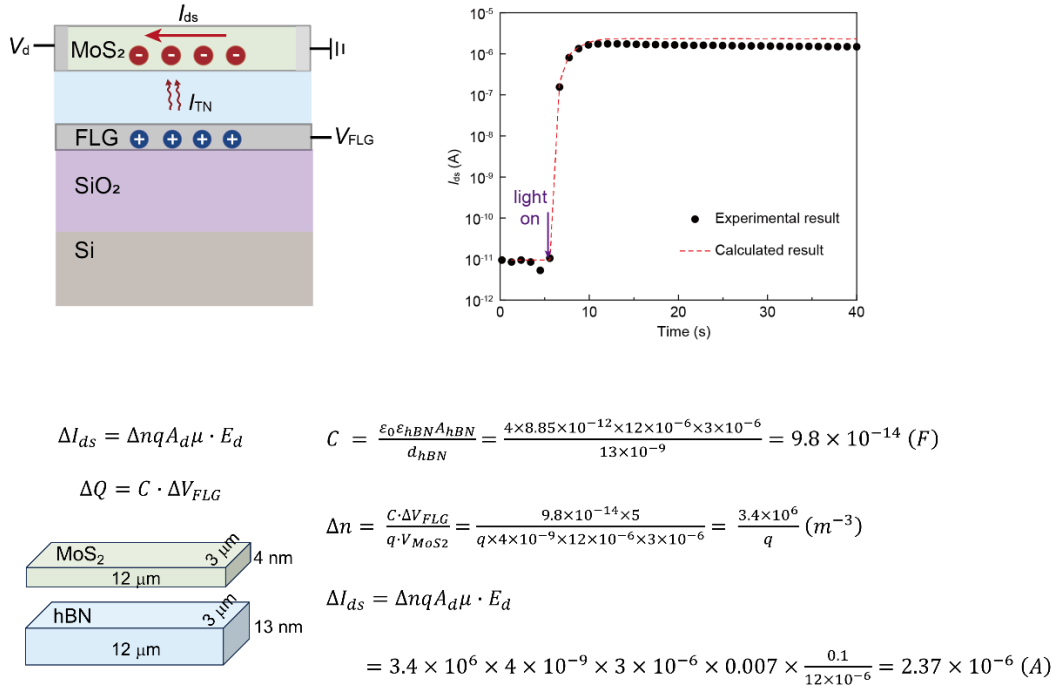


Fig. R1. The schematic structure of the device and the calculation procedure to estimate the channel current.

3. It is unclear why previous experimental data were removed. Was the data deemed unreliable? The update of the packaging method only shows a limited improvement in the device's response time. Your paper presents a large amount of data indicating that the device response time is in the second range, which is slower than common detectors by almost three orders of magnitude. Please provide an explanation for why packaging can enhance the response speed and whether the artificially set compensation current value is appropriate for achieving faster response.

→(1) The data previously showed in **Fig. 4b** were not deemed unreliable, but they have been updated by the results measured for 254 nm light. Both the previous and current version of **Fig. 4b** show the relationship response time, photocurrent, and light intensity. During the revision, we emphasized the ability to achieve a fast photoresponse time for 254 nm UV light by setting compliance current. There is no doubt that both set of data are reliable, they were measured under different wavelength of light.

(2) Packaging the detector with lens is a common approach in UV photodetectors to improve the sensitivity to weak light. In our work, since the response time is inversely proportionally to the power density of incident light, packaging the device with a quartz lens could increase the effective power density of the UV light hitting the device, leading to a faster response time.

(3) The use of a compensation current is a well-established method in electronic devices, such as in memristors. In this device, by setting a lower compliance current, the response speed is enhanced. Moreover, since we determine the light intensity based on the photoresponse time, rather than the saturation photocurrent, the setting of the compliance current does not affect the calibration of light intensity.

4. There are labeling errors in Figure S18. Moreover, the device noise is close to the lowest testing limit of the equipment, and it is necessary to provide data for the system's base noise. For the detectivity results, it is essential to include the effective area of the device.

→ Thanks for pointing out the typo in caption of Fig. S18. We have corrected this error in the revised version.

Regarding the device noise and limit of the instrumental testing limit, we acknowledge that determine the exact noise testing limit is challenging. However, we have measured the noise spectral density of another two-terminal photoconductive device based on a highly insulating material, which shows a lower noise level than the MoS₂/hBN/FLG device, as shown in **Fig. R2**. This comparison suggests that the measured noise level in our device is still above the instrument's limit. Additionally, if the actual noise level were below the instrument's limit, the detectivity would have been underestimated.

For the detectivity results, we have calculated the specific detectivity using

the formula $D^* = (R \cdot \sqrt{A})/i_n$, where the effective area of the device has been included during the calculation. Note that the effective area is the overlapping area between MoS₂ and FLG layer in this device as demonstrated in Supplementary Fig. 19.

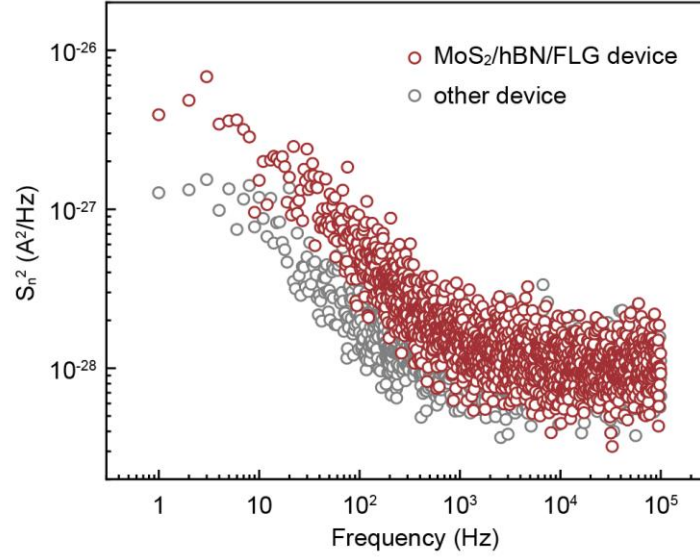


Fig. R2. Noise spectra of the MoS₂/hBN/FLG device (red) and other device (grey).

5. It is important to present the UV-visible absorption spectra for materials like graphene and MoS₂ used as absorption layers, as this would provide further insights into the device's performance.

→Thank you for your valuable suggestion. To address your concern, we have measured the UV-visible absorption spectra of both the FLG and MoS₂ layers, with thicknesses comparable to those used in the MoS₂/hBN/FLG device. The results are presented in **Fig. R3**.

As shown, FLG exhibits a broad absorption spectrum spanning the UV to visible range, with absorption increasing as the wavelength decreases. In contrast, the MoS₂ layer demonstrates a lower absorption rate compared to FLG. These findings further reinforce the conclusion that the FLG layer plays a dominant role in the UV photoresponse of the device.

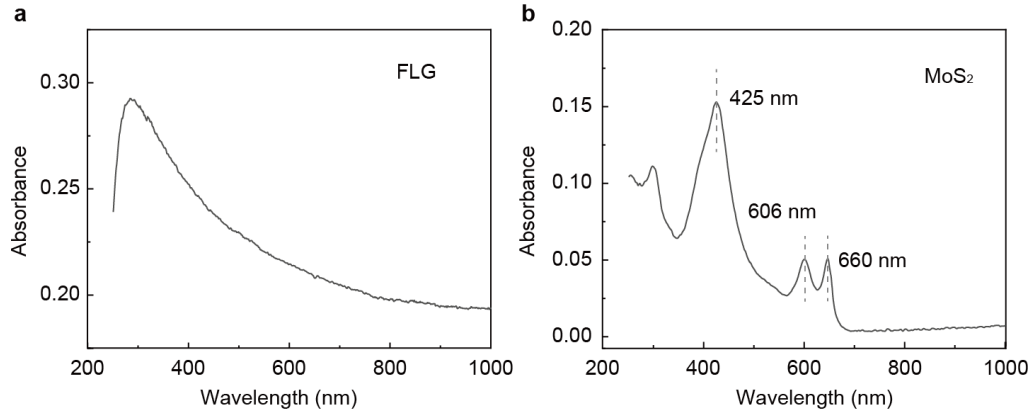


Fig. R3. The photon absorption of the FLG (a) and MoS₂ (b).

6. Generally, the generation of photogenerated carriers is not directional, the mechanism driving the effective separation of photogenerated carriers and the spontaneous tunneling of individual carriers into the channel layer remains unclear. A detailed explanation of this process is crucial for understanding the functioning of the device.

→ We thank the reviewer's question. Firstly, in this device, the channel current is not directly attribute to the transport of the photogenerated electrons or holes in MoS₂ layer. Instead, the channel current is controlled by the carrier density of MoS₂ layer. Since the MoS₂ layer is an n-type semiconductor, the channel current is dominated by electrons.

The photon excitation occurs in the absorption layer, where it facilitates the tunneling of the stored electron in the FLG layer through the hBN layer. Importantly, only one type of carrier (either electrons or holes) is dominant in the FLG layer, and this is controlled by the applied gate voltage (V_g). When a positive V_g is applied, electrons are the dominant carriers, while a negative V_g results in holes being the dominant carriers. Thus, photon excitation assists in the tunneling of only the dominant carriers (electrons or holes) during the process.

7. According to the data presented in Figure 4c, the device's responsivity linearly changes with incident light intensity over a wide range. However, you suggest that interface defects have no impact on the device performance, which contradicts the data showing significant defects. Stronger evidence and explanation are required to clarify this discrepancy.

→ Thanks for the question. The variation in photoresponsivity with incident light intensity, as shown in Figure 4c, is not due to interface defects. Instead, it arises from the nearly constant photocurrent observed across a wide range of light intensities (the underlying reason has been mentioned in response to question 2). The photoresponsivity (R) is defined as the ratio of photocurrent (I_{ph}) to the incident

light power on effective device area (P), and since the photocurrent remains almost constant for varying light intensity, the responsivity follows an inverse proportionality to the light intensity ($R \propto P^{-1}$). This behavior is consistent with the experimental data and aligns with our theoretical predictions.

To further support this explanation, we present additional data in Fig. R4, where the photocurrent remains almost constant as the light intensity varies, and the responsivity is inversely proportional to light intensity, $R \propto P^{-1}$. These results demonstrate that the observed trend in photoresponsivity is not influenced by interface defects but rather by the nature of the photocurrent's dependence on light intensity.

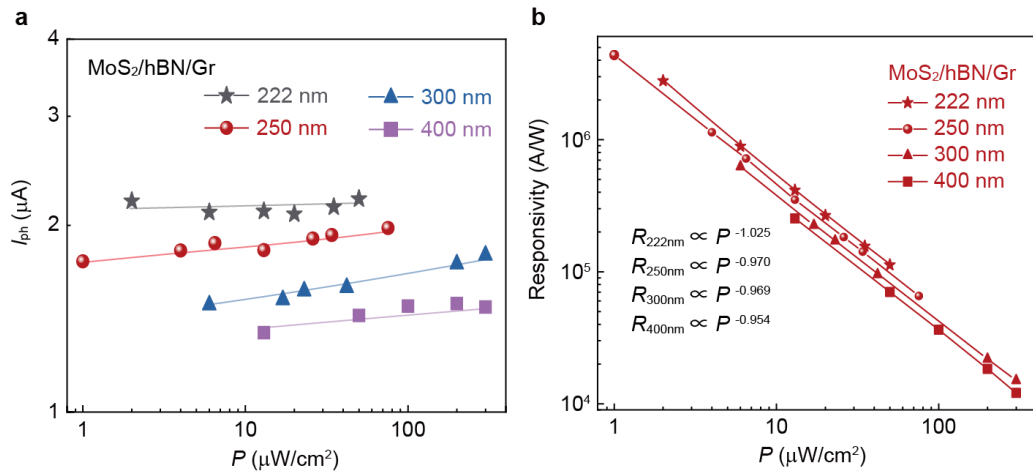


Fig. R4. Photocurrent (a) and Photoresponsivity (b) of MoS₂/hBN/FLG devices at varying wavelengths.

8. In Figure R3/R5, if the absorption layer does not respond to light in the 254-365 nm wavelength range, then it seems that the absorption layer does not contribute to the photoelectric response. Furthermore, the photogenerated current does not appear to come from the absorption layer or the channel layer. A clear explanation of where the photocurrent originates is necessary.

→ (1) As shown in previous Fig. R3, the MoS₂ layer exhibits a weak photoresponse to UV light, even though the dark current is significantly suppressed. This weak photoresponse is much lower than the large photocurrent observed in this MoS₂/hBN/FLG device. Based on this observation, we conclude that the intrinsic photoelectric effect of the MoS₂ layer has a negligible impact on the UV photoresponse. This further supports our proposed barrier-controlled photoresponse mechanism, where the primary photoresponse is governed by the tunneling barrier rather than the direct photoelectric effect of the MoS₂ layer.

(2) In Fig. R5, we also present the photoresponse of the FLG layer, where no observable photocurrent is detected. However, this lack of detectable photocurrent does not imply that the FLG layer does not absorb photons. The FLG layer exhibits

a high dark current, which may obscure any small photocurrent that could be generated under illumination. Despite this, photon absorption in the FLG layer does contribute to the device's overall photoresponse.

(3) In this device, the photon absorption in the FLG layer excites electrons, which form the non-thermal induced hot electrons. These hot electrons could easily tunnel through the hBN barrier layer, removing the stored charges in the FLG layer. and thereby altering the channel current, as discussed in response to question 2.

9. Due to these significant unresolved issues, we cannot recommend your manuscript for publication at this time. We encourage you to address these concerns in a more comprehensive manner and re-submit your work after further revisions.

→ We hope the reviewer's concern could be removed after reading the responses above. Specifically, we hope the underlying photoresponse mechanism now gets clear for the reviewer. We are glad to hear from the reviewer if there is anymore questions.