nature portfolio

Peer Review File

Transition-Metal-Doped WSe2 Homojunctions for Multifunctional Logic and Optoelectronic Devices

Corresponding Author: Professor Ngoc Thanh Duong

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

** Please ensure you delete the link to your author homepage in this email if you wish to forward it to your coauthors **

Dear Professor Duong,

Thank you again for submitting your manuscript "Transition-Metal-Doped WSe2 Homojunctions: A Unified Building-Block Platform for Emerging Electronic and Optoelectronic Devices" to Communications Materials. We have now received reports from 2 reviewers and, based on their comments, we have decided to invite a revision of your work. You will find the reviewers' reports below. While they find your work of interest, they have raised important points which must be addressed in a revised manuscript.

We recommend that you use the following title: "Transition-Metal-Doped WSe2 Homojunctions for Multifunctional Logic and Optoelectronic Devices".

To allow us to move forward with your work, we also ask that you edit your manuscript according to the table linked below. **Please read this document carefully as we will be unable to further assess your revised paper until these important points are addressed.**

Table Link:

https://springernature-my.sharepoint.com/:w:/p/vau0656/IQCDIrtHjIOXQ5kz8wf4F9jfAZKYvVvHYA8MemB8bKbTUEE?e=3KyQo8

Please outline all revisions made in the right-hand column and return the completed table with your updated manuscript files as a Related Manuscript file.

When resubmitting, please also include:

- A point-by-point response to the reviewers' comments. If you are unable to address specific reviewer requests or find any points invalid, please explain
- A clean version of your revised manuscript with no mark-ups
- A marked-up version of your paper with all changes highlighted in a different colour

Please use the link below to submit your revised files:
Link Redacted

** This url links to your confidential home page and associated information about manuscripts you may have submitted or that your are reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to receive your revised paper within six weeks, but we understand that the revisions may take longer. Please let us know if you find that the revision process will take substantially more time.

We are committed to providing a fair and constructive peer-review process. Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Kai-Qiang Lin, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0001-9609-749X

Reviewers' comments:
Reviewer #1 (Remarks to the Author):

The manuscript titled “Transition-Metal-Doped WSe2 Homojunctions: A Unified Building-Block Platform for Emerging Electronic and Optoelectronic Devices” shows a promising technique to introduce homojunction WSe2 electronics by exploiting intrinsic WSe2 with vanadium (V) doping. A group of intrinsic and doped FET devices is presented. The topic of achieving stable, uniform, and controllable p-type doping is quite critical in the field of 2D materials. Addressing the following major comments would strengthen the manuscript before publication.
1. How does the increase in V-dopant concentration correlate with the thickness of the flake in each sample? Referring to Fig. 2, below 5 nm thickness, both pristine and V-doped samples exhibit the same electrical behavior. Does it mean no effective V substitution in this range? What is the measured mass considered for the precursor calculation? Is it the assumption that the precursor decomposes completely for the chemical reaction? If complete decomposition is assumed, please state the same in the manuscript for readers and include a reference to support the assumption. Quantifying the efficiency of dopant incorporation would provide a useful metric for evaluating the effectiveness of V incorporation into WSe2 single crystals.
2. What happens to the WSe2 growth and V incorporation if salt treatment is avoided? To ensure integration with established industrial practices, this must be avoided; discuss alternatives.
3. Investigate the impact of high growth quality across the entire range of doping concentrations through quantitative electrical performance evaluation, such as mobilities, threshold voltages, and on/off ratios in each group (pristine/doped) of devices presented in Fig 2. These are essential for achieving and claiming “high-quality single crystal” growth (page no 6, line no 6) and, subsequently, for applications. A summarized table is suggested in the supporting information.
4. The transfer characteristics are measured at a relatively high drain–source bias (VDS = 3V). It would be helpful if the authors could provide transfer curves measured at lower VDS in the Supporting Information. Such data would allow verification of the intrinsic device behavior in the linear transport regime, enable a more reliable extraction of electrical parameters, confirm the consistency of the p-type transport associated with V doping in WSe2, and help rule out possible in-plane effects that may arise at higher drain biases.
5. Referring to the second device framework of the flexible photovoltaic results, include in detail the physical mechanism and the origin of the built-in electric field, which facilitates efficient photocarrier separation and collection.
6. There are some typos, like a space before the full stop on page 5, line 7, italic page 8 “(Vin), with the drain supply voltage (Vdd)”, and repeated abbreviation definition, see page 9 “behavior at VG = 0 V”, page 13, line 15, “gate voltages (VG)”. Please ensure that the corrections are made in accordance with the journal's required format.

Reviewer #2 (Remarks to the Author):

This work presents a systematic study of Vanadium-doped WSe2 homojunctions. The authors demonstrate that increasing vanadium concentration progressively enhances hole-dominant transport, which contrasts with pristine WSe2 that typically evolves toward ambipolar and n-type conduction with increasing thickness. The manuscript showcases the versatility of this doping approach through three applications: (i) complementary logic inverters, (ii) flexible photovoltaic devices, and (iii) gate-programmable in-sensor imaging devices. Given the advancement in substitutional doping control and the breadth of the device demonstrations, I recommend this work for publication in Communication Materials after the following minor revisions are addressed.
1. Could the authors clarify how the concentration of V dopant is controlled during CVT growth, and whether the transport agent or temperature gradient affects the uniformity of V distribution within the WSe2 lattice?
2. In the CVT growth of V-doped WSe2, phase purity and dopant substitution are critical issues. What experimental evidence confirms that V atoms are substitutionally incorporated into the W sites of the WSe2 lattice rather than forming secondary phases or V-related clusters?
3. Regarding the CMOS inverter, the authors should provide the individual transfer characteristics for both the NMOS (pristine) and PMOS (V-doped) transistors.
4. To support the claims regarding the fast response of the photovoltaic devices, the photocurrent response should be measured under various modulation frequencies.
5. The authors should provide data from multi-cycle photocurrent tests to confirm the mechanical durability of the device after

bending test.
6. In Figure 6b, the text describes the V-doped WSe_2 as degenerate, yet the schematic band diagram appears to represent a p-type semiconductor.
7. I recommend the authors revise the title. The term "unified building block platform" feels like an overclaim for the current scope of the work. A more descriptive title focusing on the metal-doped homojunctions and their specific applications would be more appropriate for a technical communication.

Version 1:

Decision Letter:

** Please ensure you delete the link to your author homepage in this email if you wish to forward it to your coauthors **

Dear Professor Duong,

Thank you once again for submitting your manuscript, “Transition-Metal-Doped WSe_2 Homojunctions for Multifunctional Logic and Optoelectronic Devices," to Communications Materials. Your manuscript has been seen again by Reviewer 1, whose comments are appended below. Reviewer 2 did not respond to our requests, so we checked your replies. I am happy to say that the concerns of our reviewers have now been mostly addressed, but we ask that you address the following final points before acceptance:

1. 1-50 kHz instead of 1-50 Hz.
2. Careful about stating the V concentration. Instead, it is better to state the nominal precursor ratio.
3. Strictly speaking, the authors better use "CMOS-like inverter" instead of claiming "CMOS inverter".

We also ask that you address the following editorial points:
-Please reduce the length of the abstract so that it does not exceed 180 words.
-Please add a sub-heading immediately below the 'Results and Discussion' heading.

-Please upload your main Figures files as whole files, not as individual panels (we only allow one file per figure).
Each Figure be uploaded as a separate file titled “Figure XX.” We allow only one file per Figure, with all Figure elements or panels together, displayed as intended in the final published article. Please submit your figures with one file per figure.
The above instructions are for Main figure files only and not for supplementary figures. The supplementary figures should remain in the supplementary information file as they are.
At this stage we can only accept main text figure files in the following formats: PSD, AI, EPC, TIFF, JPG, PDF, PS, GIF, PPT, PPTX, PNG, BMP, VSD, CDX, SVG, EMF, EPS. Please supply your figures following these guidelines.
Please ensure that all main text Figure captions are grouped together at the end of your main manuscript file. Embedding captions within figure files is not permitted at this stage.
Your Figure files must not include any additional text or labels that do not form part of the Figure itself.
-Please provide supplementary information file in PDF format.

This will be the final revision of your manuscript. We ask that you carefully review all files associated with your paper and follow the link below to upload the final version of all files, including display items and supplementary material. Please ensure that these files are clean, without any markups or comments, as they will be sent for publication.

Link Redacted

We hope to receive this updated version of your paper within 1 week, but please let us know if you find that you need more time.

Best regards,

Kai-Qiang Lin, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0001-9609-749X

Reviewers' comments:
Reviewer #1 (Remarks to the Author):

The authors have properly responded to all my questions and comments. I recommend this work for publication in Communications Materials.

Version 2:

Decision Letter:

Dear Professor Duong,

We are delighted to accept your manuscript titled "Transition-Metal-Doped WSe2 Homojunctions for Multifunctional Logic and Optoelectronic Devices" for publication in Communications Materials. Thank you for choosing to publish your interesting work with us.

Acceptance of your manuscript is conditional on all authors' agreement with our publication policies. In particular, your manuscript must not be published elsewhere and there must be no announcement of the work in the media until the publication date.

Please carefully read the information below for information on what to expect from the next steps of the publishing process:

Article in Press:

Please note that in advance of your paper being published we will host an early access version, known as an ‘Article in Press,’ on our journal website. For more information on this initiative please see our author guidelines.

Publication as an Article in Press is typically within 1-2 weeks after we have received your corrected proofs and publishing agreement. Subsequently, we will aim to publish the Version of Record in a timely manner. Please note there will be no further correspondence about your publication date.

When your article is published as the Version of Record, you will receive a notification email. **If you are planning an embargoed press release or require a specific publication date, please complete our scheduling requests form, or contact commsproduction@springernature.com, as soon as possible after acceptance and we will endeavour to accommodate your request.**

For further information on the journey of your article from acceptance to publication, please see our Author FAQs.

Publishing Agreements and Fees:

In about one week, you will receive an email with a link to complete the appropriate grant of rights necessary for publishing your paper and – if applicable – to provide payment information for your article-processing charge (APC), either via credit card or by requesting an invoice.

If needed, our Author Services team will be in touch regarding any additional information that may be required.

In order to avoid any delays, please ensure that you have emails from Springer Nature whitelisted in your mail system.

Proofs:

At this point we will also edit your manuscript to ensure that it conforms with our house style. Once you have completed the publication agreement and arranged payment, you will receive a separate email with a link to an online eProof for you to review. Please read your proof with great care to ensure that no changes have been introduced which have inadvertently altered the meaning of your paper. We suggest that you discuss the proof with your co-authors, but please ensure that only one author communicates with us and that only one set of corrections is returned via the online correction in the eProof.

The corresponding (or nominated) author is responsible on behalf of all co-authors for the accuracy of all content, including spelling of names and current affiliations.

To ensure prompt publication, your proofs should be returned within two working days. If there is any period within the next four weeks in which you won't be available, please nominate a co-author with whom we can correspond and send their contact information to us via email at commsproduction@springernature.com as soon as possible.

Please note that your Supplementary Information files are now finalised, and they will be submitted as provided for preparation for publication of the Article. Any requests to make changes will only be considered in exceptional circumstances and will result in a delay to publication.

You will not receive access to your eProof until the Licence to Publish and Article-Processing Charge steps are completed.

We welcome the submission of material for the ‘Featured Image’ section of the Communications Materials home page. Images should relate to the content of your manuscript, but do not need to be taken directly from the accepted work. Suggestions should be sent by email to commsmat@nature.com, along with the completed Licence to Publish form. Please note that images should be supplied as1400x400-pixel, in RGB. Unfortunately, we cannot promise that your suggestions will be used.

Providing great service is very important to us. We would greatly appreciate any comments you have about your experience at Communications Materials. We look forward to publishing your paper, and we hope to work with you again in the future.

Best regards,

Kai-Qiang Lin, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0001-9609-749X

*You can now use a single sign-on for all your accounts, view the status of all your manuscript submissions and reviews, access usage statistics for your published articles and download a record of your reviewing activity for the Nature Portfolio journals. Please check your account regularly and ensure that we have your current contact information.

** Visit Nature Research's author and referees' website at www.nature.com/authors for information about policies, services and author benefits**

We may promote your article on social media once it is published, so please feel free to send me the twitter handles of any authors or departments and we will be sure to tag them accordingly.

Dear Editor and Reviewers:

We sincerely thank the reviewers for their thorough evaluation of our manuscript entitled **"Transition-Metal-Doped WSe_2 Homojunctions: A Unified Building-Block Platform for Emerging Electronic and Optoelectronic Devices."**

Their insightful comments have significantly improved the quality and clarity of the manuscript. We have carefully addressed all comments and revised the manuscript accordingly. All changes are highlighted in the revised version.

Below, we provide a point-by-point response.

Response to Reviewer 1

The manuscript titled "Transition-Metal-Doped WSe2 Homojunctions: A Unified Building-Block Platform for Emerging Electronic and Optoelectronic Devices" shows a promising technique to introduce homojunction WSe2 electronics by exploiting intrinsic WSe2 with vanadium (V) doping. A group of intrinsic and doped FET devices is presented. The topic of achieving stable, uniform, and controllable p-type doping is quite critical in the field of 2D materials. Addressing the following major comments would strengthen the manuscript before publication.

Response: We thank the reviewer for carefully reading our manuscript and valuable comments. We especially appreciate the specific thoughtful questions, and your suggestions are helpful to improve detailed explanations for our manuscript. We would like to address the concern below point-by-point.

1. How does the increase in V-dopant concentration correlate with the thickness of the flake in each sample? Referring to Fig. 2, below 5 nm thickness, both pristine and V-doped samples exhibit the same electrical behavior. Does it mean no effective V substitution in this range? What is the measured mass considered for the precursor calculation? Is it the assumption that the precursor decomposes completely for the chemical reaction? If complete decomposition is assumed, please state the same in the manuscript for readers and include a reference to support the assumption. Quantifying the efficiency of dopant incorporation would provide a useful metric for evaluating the effectiveness of V incorporation into WSe_2 single crystals.

Response: We thank the reviewer for this valuable comment regarding the effective V substitution. The correlation between V-dopant concentration and flake thickness arises primarily from the growth kinetics and dopant incorporation efficiency during crystal formation. As the thickness increases, the growth duration and available volume for substitution also increase, leading to a higher probability of V atoms occupying W lattice sites. This results in an apparent increase in effective V concentration with flake thickness.

For thin flakes below ~5 nm (Fig. 2a–c), both pristine and V-doped WSe_2 exhibit nearly identical electrical characteristics. This does not indicate the absence of vanadium incorporation; rather, it suggests that the dopant concentration is below the threshold required to significantly modulate carrier transport, or that a fraction of V atoms may exist in electrically inactive configurations. We have revised the manuscript to mention it in the main manuscript.

We add the sentence to mention the relationship between the V-dopant concentration and the thickness of the flakes:

"*Increased flake thickness enhances dopant incorporation efficiency and substitution probability, leading to a higher effective V concentration in thicker WSe_2 layers.*"

-Revised Manuscript, Line 168-170

We add the sentence to elaborate on the behaviour of the flake below 5nm, which have same electrical behaviour:

Old:

"*However, achieving a perfectly uniform dopant distribution in multilayer WSe_2 remains challenging. Local variations in vanadium concentration may lead to spatially nonuniform electrical characteristics. with certain regions exhibiting stronger p-type behavior than others. This effect is evident in Fig. 2f, where devices fabricated with 16- and 21-nm thick samples exhibit nearly identical conductivities, whereas a thicker flake of 26 nm shows a markedly different conductivity. Such deviation is attributed to local nonuniformity in vanadium incorporation rather than a simple thickness effect.*"

New:

"However, achieving a perfectly uniform dopant distribution in multilayer WSe_2 remains challenging. Local variations in vanadium concentration may lead to spatially nonuniform electrical characteristics. For ultrathin flakes (<5 nm, Fig. 2a–c, V-doped WSe_2 exhibits electrical behaviour nearly similar to pristine WSe_2, indicating that the dopant concentration is below the threshold required to significantly influence carrier transport or that a portion of the dopants remains electrically inactive. In thick flakes, with certain flakes exhibiting stronger p-type behaviour than others, this effect is evident in Fig. 2f, where devices fabricated with 16- and 21-nm thick samples exhibit nearly identical conductivities, whereas a thicker flake of 26 nm shows a markedly different conductivity."

-Revised Manuscript, Line 170-177

Regarding precursor calculation, the measured mass corresponds to the initial mass of the precursors introduced into the growth system. In our estimation of dopant concentration, we assume that the precursors undergo complete thermal decomposition and full participation in the reaction process. This is a common approximation in vapor-phase growth methods to estimate nominal doping levels.

We agree that this assumption should be explicitly stated. Accordingly, we have revised the manuscript at the method section, to clarify that:

New:

"The dopant concentration is calculated based on the initial precursor mass under the assumption of complete decomposition and incorporation into the reaction, following established practices in TMDs growth studies.[52, 53]"

-Revised Manuscript, Line 368-370

In addition, the actual doping concentration is evaluated by electron probe microanalyzer (EPMA) method (as shown in Fig. R1). The data was reported in Supplementary Information Fig. 1 in our previous report (Phys. Rev. B. 103, 014441 (2021)). The actual doping concentration is approximately two to three times smaller than the nominal values in bulk.

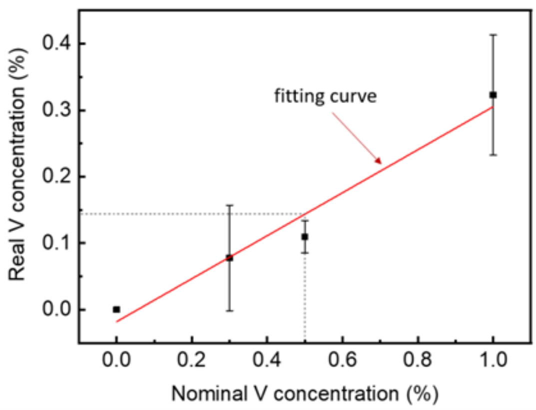


Figure R1. V doping concentration in bulk evaluated by EPMA method (Phys. Rev. B. 103, 014441 (2021)).

We acknowledge that the actual dopant incorporation efficiency may deviate from the nominal value due to factors such as incomplete precursor decomposition, parasitic reactions, or limited substitution probability. This is a reason why we have included a discussion on dopant incorporation efficiency for thin-layer samples in the manuscript, in which the advanced atomic characterization such as XPS, ADF-STEM techniques are reported to experimentally quantify the V concentration (Fig. 1 and Supplementary Fig. 2). This provides a more accurate metric for evaluating the effectiveness of V incorporation into WSe_2 single crystals.

2. What happens to the WSe_2 growth and V incorporation if salt treatment is avoided? To ensure integration with established industrial practices, this must be avoided; discuss alternatives.

Response: We thank the reviewer for this discussion. We would like to clarify that, in this work, V-doped WSe_2 single crystals are synthesized using a chemical vapor transport (CVT) method, which inherently does not rely on salt-assisted growth.

In contrast to CVD processes, where alkali salts are often introduced to enhance precursor volatility and mass transport, CVT growth utilizes transport agents (as we used I_2) to form volatile metal-halide species. This enables efficient and controlled transport of both W and V species in the vapor phase without the need for alkali salts. As a result, high-quality single-crystal WSe_2 with effective V incorporation can be achieved under salt-free conditions.

If salt treatment were avoided in conventional CVD growth, one would typically observe reduced precursor transport, leading to smaller domain sizes and significantly suppressed dopant incorporation due to the lack of volatile intermediate species. However, in the CVT approach employed here, these limitations are mitigated by the transport chemistry itself. The formation of volatile tungsten and vanadium halides ensures sufficient mass transport and promotes substitutional incorporation of V into the WSe_2 lattice during crystal growth.

We agree with the reviewer that avoiding alkali salts is essential for compatibility with industrial semiconductor processes due to contamination concerns. In this work, CVT represents a viable salt-free synthesis route for bulk single crystals. For scalable thin-film growth, alternative strategies such as metal–organic chemical vapor deposition (MOCVD) using volatile precursors (e.g., $W(CO)_6$ and VCl_x), hydrogen-assisted growth, or plasmaenhanced CVD can be employed to achieve efficient growth and controlled doping without introducing alkali contaminants.

We revise the manuscript to elaborate on issues:

Old:

The pristine and V-doped WSe2 single crystals were synthesized by a two-step chemical vapor transport (CVT) strategy, in which a controlled temperature gradient drives material transport from the hot zone to the cold zone, resulting in high-quality single crystals, as illustrated in Fig. 1a and described in the Method section.

New:

The pristine and V-doped WSe2 single crystals were synthesized by a two-step chemical vapor transport (CVT) strategy, in which a controlled temperature gradient drives material transport from the hot zone to the cold zone. I2 was employed as the transport agent, enabling efficient vapor-phase transport of both W and V species without the need for salt treatment. The salt-free approach yields high-quality single crystals, as illustrated in Fig. 1a and described in the Method section.

-Revised Manuscript, Line 122-126

3. Investigate the impact of high growth quality across the entire range of doping concentrations through quantitative electrical performance evaluation, such as mobilities, threshold voltages, and on/off ratios in each group (pristine/doped) of devices presented in Fig 2. These are essential for achieving and claiming "high-quality single crystal" growth (page no 6, line no 6) and, subsequently, for applications. A summarized table is suggested in the supporting information.

Response: We thank the reviewer for this important suggestion. In response, we have now performed a comprehensive quantitative analysis of electrical performance across all doping concentrations and flake thicknesses (Supplementary Fig. 10), including essential parameters, field-effect mobility (μ_{EF}), on-off ratio, and threshold voltage (V_{th}).

We have added further discussion on to the revised manuscript:

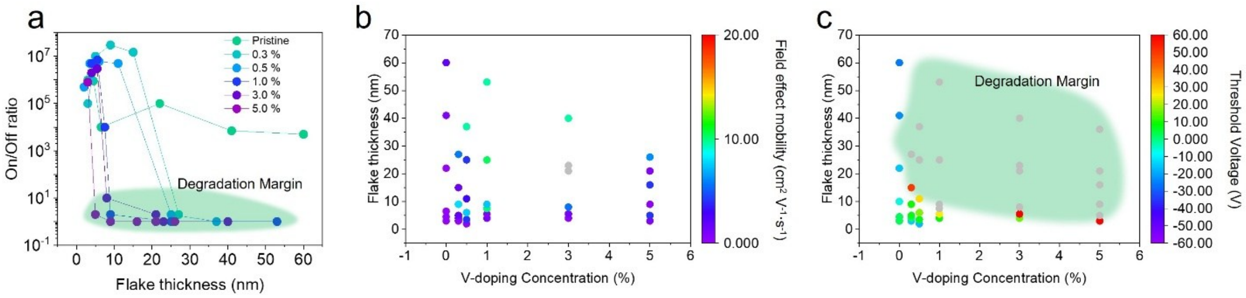
New:

"To quantitatively evaluate the electrical performance of V-doped WSe_2, we systematically extracted the field-effect mobility (μ_{FE}), threshold voltage (V_{th}), and on/off current ratio across a wide range of flake thicknesses and vanadium concentrations, as summarized in Supplementary Fig. 10. The results reveal a clear doping-dependent transport behavior. Pristine WSe_2 exhibits relatively low mobility (<2 $cm^2\ V^{-1}\ s^{-1}$) and strong thickness dependence, while moderate V doping (0.5–1%) significantly enhances carrier transport, yielding peak mobilities of ~10 $cm^2\ V^{-1}\ s^{-1}$ with minimal thickness sensitivity. Concurrently, the on/off ratio improves at low-to-moderate doping (up to ~10^7–10^8) but degrades at higher doping concentrations (≥ 3 %) due to increased off-state leakage and reduced gate modulation. The threshold voltage systematically shifts toward more positive values with increasing V concentration, confirming effective p-type doping via substitutional vanadium incorporation. At higher doping levels, the devices approach degenerate behavior, resulting in diminished switching characteristics and less well-defined threshold voltages. These combined trends identify an optimal doping window (~0.5–1 %), where enhanced mobility, high on/off ratio, and stable threshold behavior are simultaneously achieved, highlighting the robustness and tunability of V-doped WSe_2 for electronic and optoelectronic applications."

-Revised Manuscript, line 178-192

We have also added Supplementary Figure 10. with additional discussion:

New:



Supplementary Figure 10. *(a) On/off current ratio as a function of flake thickness for pristine and V-doped* WSe_2 *devices with varying vanadium concentrations. Moderate doping (0.3–0.5%) enhances switching performance, while higher doping (≥3%) leads to degraded gate modulation and reduced on/off ratios, approaching degenerate behavior. The shaded region indicates the degradation regime. (b) Field-effect mobility (μFE) mapped as a function of flake thickness and V-doping concentration. Moderate doping (0.5–1%) yields enhanced and thickness-tolerant mobility (~8–10* $cm^2\ V^{-1}\ s^{-1}$*), whereas higher doping levels introduce increased variability due to impurity scattering and non-uniform dopant distribution. (c) Threshold voltage (*V_{th}*) extracted using a constant-current method (*I_D*= 1 nA), as a function of thickness and doping concentration. A systematic positive shift in Vth with increasing V concentration confirms effective p-type doping, while highly doped devices exhibit reduced gate tunability and less well-defined threshold behavior.*

We systematically extracted key electrical parameters, including field-effect mobility (μ_{FE}*), threshold voltage (*V_{th}*, defined at* $I_D = 1nA$*), and on/off current ratio for all devices shown in Fig. 2. The mobility was calculated using* $\mu_{FE} = (L/WC_{ox}V_{DS})(dI_D/dV_G)$*, where* $L = 1\ \mu m$*,* $W = 2\ \mu m$*, and* C_{ox}*corresponds to 300 nm* SiO_2*. As summarized in Supplementary Fig. 10, the results reveal a clear thickness- and doping-dependent transport behavior. Pristine* WSe_2 *exhibits relatively low mobility (<2* $cm^2\ V^{-1}\ s^{-1}$*) and strong thickness dependence, while moderate V doping (0.3–0.5%) enhances switching performance, yielding high on/off ratios (~*10^7*–*10^8*). Further increasing the doping concentration improves mobility, reaching ~8–10* $cm^2\ V^{-1}\ s^{-1}$ *for 0.5–1% doping with minimal thickness dependence, indicating effective and uniform substitutional doping. However, higher doping levels (≥3%) lead to reduced on/off ratios and poorly defined threshold voltages due to increased off-state leakage and degenerate transport behavior. The threshold voltage systematically shifts toward more positive values with increasing V concentration, confirming p-type doping. Overall, these results identify an optimal doping window (~0.5–1%) that balances high mobility, strong gate modulation, and stable threshold behavior. The consistent trends across multiple devices further confirm the high crystalline quality and reliable dopant incorporation in V-doped* WSe_2*, while extracted values represent apparent mobility and exclude outliers arising from non-linear transport regimes.*

4. The transfer characteristics are measured at a relatively high drain–source bias (V_{DS} *= 3V). It would be helpful if the authors could provide transfer curves measured at lower* V_{DS} *in the Supporting Information. Such data would allow verification of the intrinsic device behavior in the linear transport regime, enable a more reliable extraction of electrical parameters, confirm the consistency of the p-type transport associated with V doping in* WSe_2*, and help rule out possible in-plane effects that may arise at higher drain biases.*

Response: We thank the reviewer for this insightful comment. To address this concern and to verify the intrinsic transport behavior in the linear regime, we have additionally measured the transfer characteristics at a lower drain bias of V_{DS} = 1 V, (**Figure 2**). The results show consistent electrical behavior with those obtained at V_{DS}= 3 V, including the preserved p-type transport characteristics in V-doped WSe_2 and the overall trend of thickness- and doping-dependent modulation.

We have added the description to the revised manuscript:

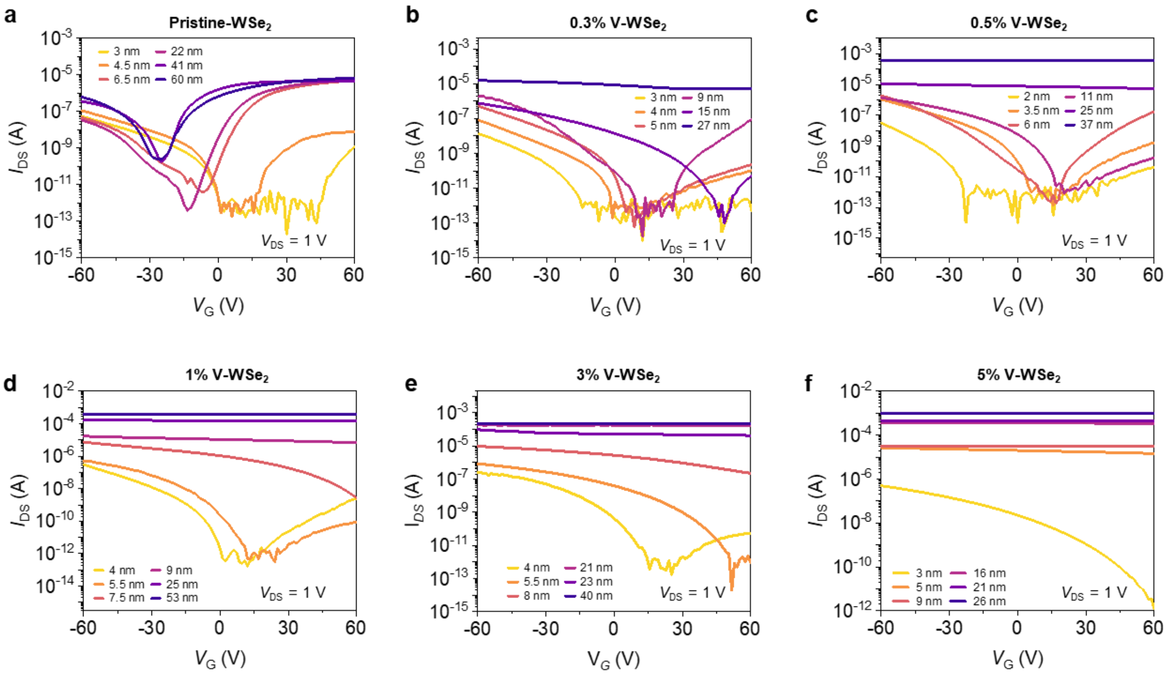
New:

"The transfer characteristics measured at a lower drain bias (V_{DS} = 1 V), as shown in Supplementary Fig. 9, exhibit behavior consistent with those obtained at V_{DS} = 3 V."

-Revised Manuscript, Line 151-153

We have also added the new Supplementary Figure 9:

New:



Supplementary Figure 9. *Thickness-dependent transfer characteristics of pristine and V-doped WSe_2 (V_{DS} = 1 V). (a) Pristine WSe_2 devices exhibit a transition from p-type-dominant to n-type-dominant transport with increasing thickness. (b–f) V-doped WSe_2 (0.3–5%) shows suppressed ambipolarity and progressively enhanced p-type conduction with increasing dopant concentration, which is further strengthened in thicker layers, indicating thickness-tolerant p-type modulation. The transfer characteristics measured at a lower drain bias (V_{DS} = 1 V) confirm consistent behavior with those obtained at higher bias (V_{DS} = 3 V).*

5. Referring to the second device framework of the flexible photovoltaic results, include in detail the physical mechanism and the origin of the built-in electric field, which facilitates efficient photocarrier separation and collection.

Response: We thank the reviewer for this valuable comment. To further clarify the physical mechanism and the origin of the built-in electric field in the flexible photovoltaic homojunction, we have expanded the discussion based on the KPFM results (Figure 4d, e). The work-function (WF) difference between pristine WSe_2 and 0.5% V-doped WSe_2 can be estimated from the measured contact potential difference (CPD) using the relation, $\varnothing_{Sample} = \varnothing_{Tip} - \Delta_{CPD}$, where $\varnothing_{Sample}$ and $\varnothing_{Tip}$ are the WF of the sample and WF of the tip, respectively) [*Nat Commun* 7, 13413 (2016)]. According to this relation, a lower measured surface potential corresponds to a higher sample WF.

As shown in Supplementary Fig. 13a, before contact, the CPD of the 0.5% V-doped WSe_2 region is approximately 0.38 eV lower than that of pristine WSe_2. This indicates that V doping increases the WF of WSe_2 and shifts its Fermi level downward toward the valence band, consistent with enhanced p-type behavior. When the pristine and V-doped regions are brought into contact, charge redistribution occurs across the interface until the Fermi levels become aligned. This equilibration results in band bending and establishes a built-in electric field at the homojunction interface, as illustrated in Supplementary Fig. 13b. Under light illumination, this internal electric field efficiently separates photogenerated electron-hole pairs, driving electrons toward the pristine WSe_2 side and holes toward the 0.5% V-doped WSe_2 side, as shown in Fig. 5d. These results explain the photovoltaic behavior of the flexible homojunction device and confirm that the built-in electric field originates from the V-induced WF difference between the two WSe_2 regions.

We have added the description to the revised manuscript:

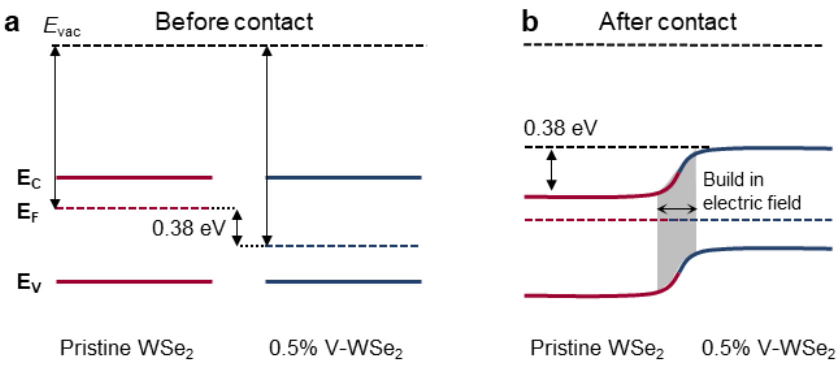
New:

"To directly probe the formation of a built-in potential at the 0.5% V-doped / pristine WSe_2 interface, KPFM measurements were performed, as shown in Figs. 4d. The work-function (WF) difference between pristine WSe_2 and 0.5% V-doped WSe_2 can be estimated from the measured contact potential difference (CPD) using the relation: $\varnothing_{Sample} = \varnothing_{Tip} - \Delta_{CPD}$, where $\varnothing_{Sample}$ and $\varnothing_{Tip}$ are the WF of the sample and WF of the tip, respectively) [Nat Commun 7, 13413 (2016)]. According to this relation, a lower measured surface potential corresponds to a higher sample WF. As shown in Fig 4e and Supplementary Fig. 13, before contact, the CPD of the 0.5% V-doped WSe_2 region is approximately 0.38 eV lower than that of pristine WSe_2. This indicates that V doping increases the WF of WSe_2 and shifts its Fermi level downward toward the valence band, consistent with enhanced p-type behavior. When the pristine and V-doped regions are brought into contact, charge redistribution occurs across the interface until the Fermi levels become aligned. This equilibration results in band bending and establishes a built-in electric field at the homojunction interface."

-Revised Manuscript, Line 230-241

We have also added the new Supplementary Figure 13:

New:



Supplementary Figure 13. *Schematic energy band diagrams of pristine WSe_2 and 0.5% V-WSe_2 derived from KPFM measurement. (a) before contact, showing the work-function difference between pristine and doped WSe_2; (b) after contact, showing Fermi-level alignment, interfacial band bending, and formation of the built-in electric field.*

6. There are some typos, like a space before the full stop on page 5, line 7, italic page 8 "(V_{in}), with the drain supply voltage (V_{dd})", and repeated abbreviation definition, see page 9 "behavior at V_G = 0 V", page 13, line 15, "gate voltages (V_G)". Please ensure that the corrections are made in accordance with the journal's required format.

Response: We thank the reviewer for the invaluable feedback provided by the reviewer, which has greatly contributed to the refinement of our manuscript. As per your guidance, we have checked and modified substantial revisions in multiple sections to enhance the manuscript's quality and in accordance with the journal's guidelines.

Response to Reviewer 2

This work presents a systematic study of Vanadium-doped WSe_2 homojunctions. The authors demonstrate that increasing vanadium concentration progressively enhances hole-dominant transport, which contrasts with pristine WSe_2 that typically evolves toward ambipolar and n-type conduction with increasing thickness. The manuscript showcases the versatility of this doping approach through three applications: (i) complementary logic inverters, (ii) flexible photovoltaic devices, and (iii) gate-programmable in-sensor imaging devices. Given the advancement in substitutional doping control and the breadth of the device demonstrations, I recommend this work for publication in Communication Materials after the following minor revisions are addressed.

Response: We thank the reviewer for carefully reading our manuscript and valuable comments. We especially appreciate the specific thoughtful questions, and your suggestions are helpful to improve detailed explanations for our manuscript. We would like to address the concern below point-by-point.

1. Could the authors clarify how the concentration of V dopant is controlled during CVT growth, and whether the transport agent or temperature gradient affects the uniformity of V distribution within the WSe_2 lattice?

Response: We thank the reviewer for this insightful comment regarding the control of V incorporation during the CVT growth process.

In our growth procedure, the concentration of V dopant is primarily controlled by the initial stoichiometric ratio of the precursor materials used in the sealed quartz ampoule. Specifically, the starting powders of W, Se, and V were weighed according to the desired nominal composition prior to the growth process, following the nominal $V_xW_{1-x}Se_2$ (x is the V doping concentration). Because the chemical vapor transport method proceeds through equilibrium between the source and growth zones, the final dopant incorporation generally reflects the relative chemical potential of the elements in the precursor mixture.

The transport agent Iodine (halogen-based transport agents) also influences the transport kinetics and may affect the relative transport rates of W- and V-containing species. However, under the optimized growth conditions used in this study, the concentration of the transport agent was kept sufficiently low to ensure controlled and stable transport while minimizing compositional fluctuations.

Additionally, the temperature gradient between the source and growth zones plays an important role in determining the transport rate and crystal growth dynamics. A carefully optimized gradient was employed to maintain steady supersaturation conditions, which helps promote homogeneous incorporation of V atoms during crystal formation.

2. In the CVT growth of V-doped WSe_2, phase purity and dopant substitution are critical issues. What experimental evidence confirms that V atoms are substitutionally incorporated into the W sites of the WSe_2 lattice rather than forming secondary phases or V-related clusters?

Response: We appreciate the reviewer's important comment regarding phase purity and the substitutional incorporation of V dopants. To verify that V atoms are incorporated into the WSe_2 lattice rather than forming secondary phases or V-related clusters, several complementary characterization techniques were employed:

- **X-ray diffraction (XRD)** analysis shows that all diffraction peaks can be indexed to the hexagonal crystal structure of WSe_2 without the appearance of additional impurity peaks that would indicate secondary phases. Moreover, a slight shift in the diffraction peak positions is observed compared with pristine WSe_2, suggesting lattice perturbation associated with V incorporation (Fig. 1).

- **Raman spectroscopy** measurements reveal characteristic vibrational modes consistent with the host WSe_2 structure, while no additional Raman modes associated with possible vanadium selenide compounds are detected (Fig. 1 and Supplementary Fig. 1).

- **X-ray Photolumenesence Spectroscopy (XPS)** measurement displays the V doping presence in the host WSe_2 by V core peak (Fig. 1).

- **Annular Dark-field Scanning Transmission Electron Microscopy (ADF-STEM)** reveals the actual concentration at nanoscale of nominal V-doped sample (Fig. 1 and Supplementary Fig. 2). The actual V concentration is approximately 0.41%, quite close to the nominal doping concentration (~0.5%).

Finally, the relatively small ionic radius difference between V and W makes substitutional doping at the W site energetically favorable, which is consistent with previously reported studies of transition-metal-doped WSe_2 systems. Taken together, these structural and compositional analyses strongly support the conclusion that V atoms are primarily incorporated substitutionally into the W sites of the WSe_2 lattice rather than forming secondary phases.

The corresponding explanations and supporting discussions have been clarified on Supplementary Fig. 2.

3. Regarding the CMOS inverter, the authors should provide the individual transfer characteristics for both the NMOS (pristine) and PMOS (V-doped) transistors.

Response: We thank the reviewer for the constructive comment. In response, we have now included the individual transfer characteristics of both the NMOS (pristine WSe_2) and PMOS (0.5% V-doped WSe_2) as shown in Supplementary Fig. 11. These data clearly confirm the n-type behavior of pristine WSe_2 and the p-type behavior of 0.5% V-doped WSe_2.

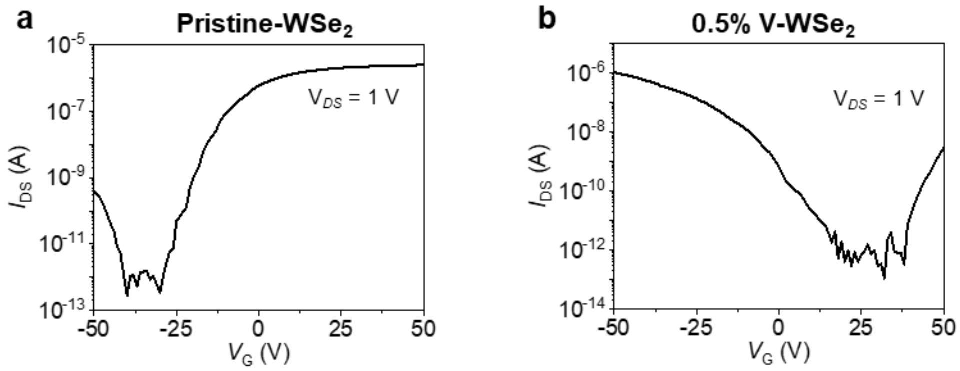
We have added the description to the revised manuscript:

New:

"The transfer curves of the individual pristine and V-doped WSe_2 devices are shown in Supplementary Fig. 11."

-Revised Manuscript, Line 198-199

We have also added the new Supplementary Figure 11:



Supplementary Figure 11. *Transfer characteristics of pristine and 0.5% V-doped WSe_2, respectively. The pristine WSe_2 exhibits n-type transport behavior, whereas the V-doped WSe_2 shows p-type transport behavior.*

4. To support the claims regarding the fast response of the photovoltaic devices, the photocurrent response should be measured under various modulation frequencies.

Response: We sincerely thank the reviewer for this valuable suggestion. To further support our result of fast photoresponse, we have performed frequency-dependent photocurrent measurements over a modulation range from 1 kHz to 50 kHz (Supplementary Fig. 4). The device exhibits stable and consistent photocurrent amplitudes across the investigated frequency range, indicating minimal signal degradation at higher modulation frequencies. These results confirm the reliability of the extracted response times and demonstrate the capability of the device for high-speed operation.

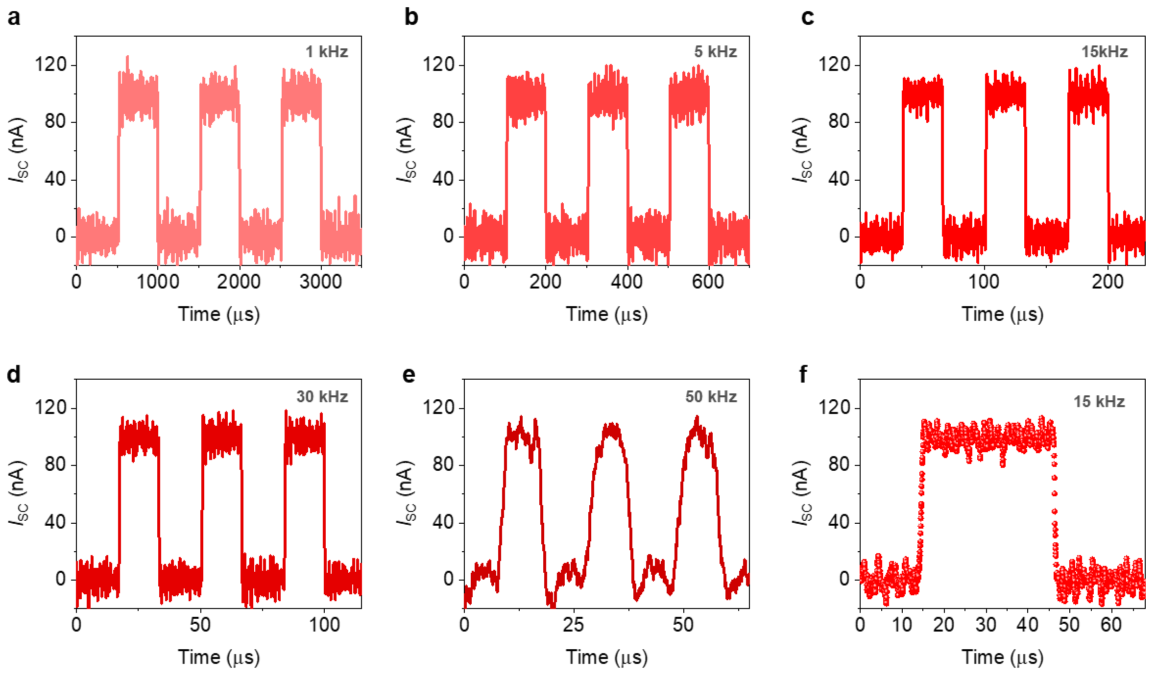
We have added the description to the revised manuscript:

New:

"*The frequency-dependent Iph response of the device over 1-50 Hz (Supplementary Fig. 17) shows stable, consistent Iph amplitudes across all measured frequencies.*"

-Revised Manuscript, Line 297-299

We have also added the new Supplementary Figure 17:



Supplementary Figure 17*. Frequency-dependent photocurrent response of the device. (a–e) Time-resolved photocurrent measured under modulated illumination with frequencies ranging from 1 to 50 kHz, showing stable and nearly constant photocurrent amplitudes of ~ 100 nA across the entire frequency range. (f) Extracted photocurrent at 15 kHz, normalized to evaluate the fast response characteristics.*

5. The authors should provide data from multi-cycle photocurrent tests to confirm the mechanical durability of the device after bending test.

Response: We sincerely thank the reviewer for this valuable suggestion. In response, we have performed multi-cycle photocurrent measurements before and after the bending tests (Supplementary Fig. **16)**. The device exhibits stable and reproducible photocurrent over repeated on/off illumination cycles, with negligible variation in photocurrent amplitude after bending. This consistency confirms that the optoelectronic performance is well preserved under mechanical deformation, demonstrating the excellent mechanical robustness and durability of the flexible device.

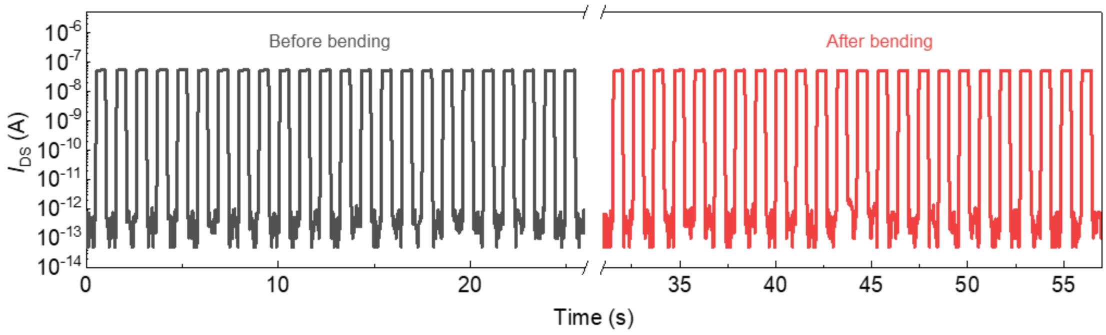
We have added the description to the revised manuscript:

New:

"Time-resolved Iph measurements at VDS = 0 V before and after bending are shown in Supplementary Fig. 16, indicating the stable switching under repeated light on/off cycles, with negligible change in Iph amplitude after mechanical deformation."

-Revised Manuscript, Line 268-270

We have also added the new Supplementary Figure 16:



Supplementary Figure 16*. Time-dependent photocurrent of the device at V_{DS} = 0 V before and after bending. The time-resolved I_{DS} under periodic light on/off cycles shows consistent photocurrent magnitude before and after bending, demonstrating the mechanical stability and reliable photoresponse of the device under deformation.*

6. In Figure 6b, the text describes the V-doped WSe_2 as degenerate, yet the schematic band diagram appears to represent a p-type semiconductor.

Response: We thank the reviewer for the invaluable feedback provided by the reviewer. We acknowledge that the original schematic in Fig. 6b did not clearly distinguish between degenerate p-type behavior and conventional p-type band representation, which may have led to confusion. In the revised manuscript, we have updated the band diagram to more accurately reflect the degenerate nature of the V-doped WSe_2 (Fig. 6b). Specifically, the Fermi level in the V-doped WSe_2 is now depicted closer to (or within) the valence band, consistent with its high hole concentration and weak gate tunability observed experimentally.

We have revised Fig. 6b as below.

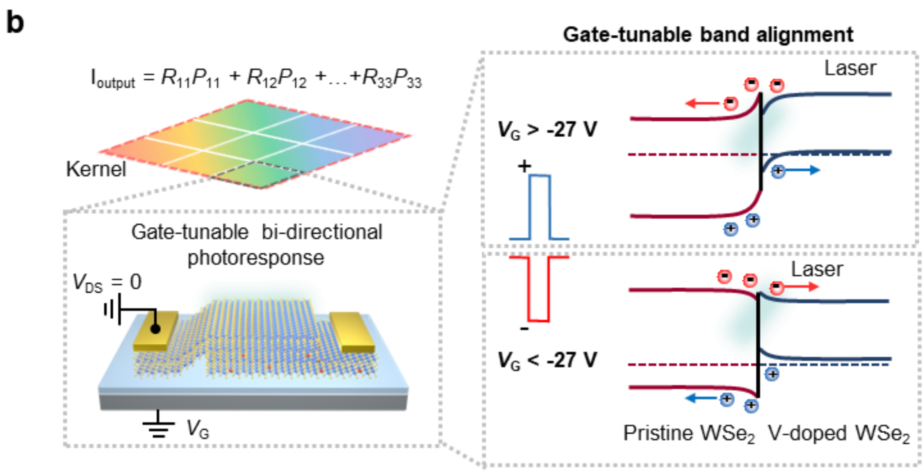


Figure 6b. Band diagram to more accurately reflect the degenerate nature of the V-doped WSe_2.

7. I recommend the authors revise the title. The term "unified building block platform" feels like an overclaim for the current scope of the work. A more descriptive title focusing on the *metal-doped homojunctions and their specific applications would be more appropriate for a technical communication.*

Response: We thank the reviewer for this valuable and constructive suggestion. Following the reviewer's recommendation, we have revised the title to more accurately reflect the key focus of our study on doped homojunctions and their demonstrated device applications.

The revised title is:

"Transition-Metal-Doped WSe_2 Homojunctions for Multifunctional Logic and Optoelectronic Devices"

Dear Editor and Reviewers:

We sincerely thank the Editor for the positive evaluation and for the additional comments regarding our manuscript. We have carefully addressed all remaining reviewer and editorial points in the revised manuscript, including correcting the frequency range to 1–50 kHz, revising the terminology to use nominal precursor ratios instead of V concentration, replacing "CMOS inverter" with "CMOS-like inverter," shortening the abstract to below 180 words, and adding a sub-heading below the "Results and Discussion" section.

We have carefully addressed all comments and revised the manuscript accordingly. All changes are highlighted in the revised version.

Below, we provide a point-by-point response.

1. We revised the abstract to reduce its length of the abstract to below 180 words:

Old:

"*Control of carrier polarity in two-dimensional van der Waals semiconductors is a fundamental requirement for the development of integrated electronic and optoelectronic systems. Here, we demonstrate that substitutional vanadium doping provides a robust route to p-type modulation in multilayer tungsten diselenide (WSe_2), resulting in pronounced thickness-dependent electronic behavior. The D.C. current-voltage characterization of V-doped transistor reveals that increasing vanadium concentration progressively enhances hole-dominant transport, in contrast to pristine WSe_2, which evolves toward ambipolar and n-type conduction with increasing thickness. Leveraging this thickness-engineered of WSe_2/V-WSe_2 material platform, we demonstrate three distinct device architectures: (i) Complementary logic inverters, (ii) flexible photovoltaic devices, and (iii) gate-programmable in-sensor imaging devices. The flexible photovoltaic homojunctions exhibit a fast photoresponse with a rise time of 1.36 μs, broadband photodetection spanning the ultraviolet to near-infrared, and a high external quantum efficiency of up to 51.3%, while maintaining stable performance* *under mechanical deformation. Separately, complementary inverter devices demonstrate reliable logic functionality enabled by controlled carrier polarity, while gate-tunable homojunction devices are engineered to reversibly switch between negative and positive photocurrent (I_{ph}) responses through gate-modulated band alignment, which is further exploited to realize programmable in-sensor image processing. Our results establish substitutional vanadium doping as an effective strategy for controlling carrier polarity in multilayer WSe_2 and enabling high-performance WSe_2/V-WSe_2 -based homojunction electronic and optoelectronic devices."*

New: [*177 words*]

"Control of carrier polarity in two-dimensional van der Waals semiconductors is essential for integrated electronic and optoelectronic systems. Here, we demonstrate that substitutional vanadium doping enables robust p-type modulation in multilayer WSe_2 with pronounced thickness-dependent transport behavior. The transfer curve of V-doped transistor reveals that increasing vanadium concentration progressively enhances hole-dominant transport, in contrast to pristine WSe_2, which evolves toward ambipolar and n-type conduction with increasing thickness. Leveraging this thickness-engineered of WSe_2/V-WSe_2 platform, we demonstrate three distinct device architectures: (i) Complementary logic inverters, (ii) flexible photovoltaic devices, and (iii) gate-programmable in-sensor imaging devices. Flexible photovoltaic homojunctions exhibit a fast response time of 1.36 μs, broadband photodetection spanning the ultraviolet to near-infrared, and a high external quantum efficiency of up to 51.3%, while maintaining stable performance under mechanical deformation. Separately, complementary inverter devices demonstrate reliable logic functionality, whereas gate-tunable homojunctions reversibly switch between negative and positive photocurrent through gate-modulated band alignment, enabling programmable in-sensor image processing. Our results establish vanadium doping as an effective strategy for carrier polarity control and for high-performance WSe_2/V-WSe_2-based homojunction electronic and optoelectronic devices."

-Revised Manuscript, Line 25-40

2. The sub-heading is added immediately below the 'Results and Discussion'

New:

“Pristine and V-doped WSe_2 growth and characterization”

-Revised Manuscript, Line 119

3. We have corrected the frequency range in the revised manuscript from 1–50 Hz to 1–50 kHz.

New:

“1-50 kHz”

-Revised Manuscript, Line 297

4. We have revised the manuscript to avoid directly stating the V concentration and clarified that the reported values refer to the nominal V: W precursor ratio used prior to growth. The following sentence has been added to the "Results and Discussion" and the "Methods " sections:

New:

“The V concentration corresponds to the nominal V: W precursor ratio used prior to growth, with the formula $V_xW_{1-x}Se_2$.”

-Revised Manuscript, Line 124-125

New:

“The V concentration denotes the nominal V: W precursor ratio before material synthesis.”

-Revised Manuscript, Line 362-363

5. We have revised the terminology throughout the manuscript to use “CMOS-like inverter” instead of “CMOS inverter” in the manuscript and Supporting information.