

Multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonic synapses

Corresponding Author: Professor Xi Chen

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 Chen,

Thank you again for submitting your manuscript "Multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonic synapses" to Communications Materials. We have now received reports from 3 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.

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

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 you 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,

Sunkook Kim, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0003-1747-4539

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The manuscript presents an intriguing approach to multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonic synapses. The work is timely and addresses an important direction in optical AI hardware. However, the conceptual basis of the synaptic weight definition and several technical details require substantial clarification. In particular, the equivalence of transmittance change (ΔT) with synaptic weight is not sufficiently justified and poses a fundamental concern. I therefore recommend major revision before further consideration.

1) Definition of Synaptic Weight (ΔT as weight):

The central issue is that the manuscript defines synaptic weight solely as the change in transmittance (ΔT). While ΔT may serve as an optical analogue to conductance modulation, its physical meaning, reproducibility, and comparability to established synaptic weights are insufficiently justified. This definition risks reducing the work to a simple demonstration of optical modulation rather than a robust synaptic weight implementation.

The authors should:

- Clarify how ΔT quantitatively maps onto weighted summation in optical neural networks.
- Discuss whether ΔT can provide stable, reproducible weight states under practical conditions.
- Benchmark ΔT -based weight against conductance-defined weights in memristive or transistor-based synapses.

Without addressing these points, the claim of "all-photonic synaptic weights" remains conceptually weak.

2) Stability and cycling data:

The manuscript provides stability tests (20 cycles, 21 days), which are relatively limited. Longer-term cycling or at least a discussion of retention, variability, and device-to-device reproducibility is needed.

3) Abrupt Optical Transitions:

The transmittance change in some plots appears abrupt, suggesting threshold-like behavior. A logarithmic scale or more detailed fitting would clarify whether the switching is analog or binary-like.

Reviewer #2 (Remarks to the Author):

The manuscript entitled "Multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonic synapses" written by Zi, Sun, and Chen et al. reports an all-photonic synapse based on photochromic dihydrated perovskites and demonstrates multidimensional as well as reconfigurable optical neuromorphic computing. The concept is interesting, and the experimental results clearly demonstrate synaptic plasticity and the high accuracy of RNN and ANN. However, there remain several points that require further clarification before the work can be considered for publication. The detailed questions are listed below.

1. Figure 3 shows that the device exhibits a markedly faster increase in transmittance during the re-learning process than during the initial learning cycle. Could the authors clarify the physical mechanism underlying this phenomenon?

2. Figure 4 presents classification tasks with relatively few classes and limited data, where one might expect even simple models to achieve high accuracy. While the RNN rapidly converges to 100%, the ANN does not. Could the authors elaborate on the factors that prevented the ANN from reaching perfect accuracy?

3. A conventional D2NN typically leverages different input modalities (e.g., distinct wavelengths or polarization states) to process and discriminate multiple datasets. However, in Figure 5, the presented "reconfigurable D2NN" seems to sequentially train and test the same network on two different datasets (MNIST and Fashion-MNIST) under identical optical conditions. From this perspective, the system appears to be more "reusable" than "reconfigurable." Could the authors clarify what distinguishes their approach as reconfigurable rather than merely sequential reuse, given that the same wavelength, humidity, and power conditions were maintained?

4. If these all-photonic perovskite synapses are to be integrated into practical hardware, how would they operate within a neural network architecture? In electronic systems, input and output neurons are represented by voltage vectors; synaptic

weights correspond to conductance values; and computation is governed by Kirchhoff's law. Similarly, how can all-photonic perovskite synapses be employed to perform neural network operations? A conceptual outline or framework for such an optical computing architecture would be highly valuable.

Reviewer #3 (Remarks to the Author):

The authors have presented the development of perovskite-based all-photonic synapses for multidimensional and reconfigurable optical neuromorphic computing. However, several key areas require further clarification and improvement:

1. The fabrication details of the synaptic device and its physical dimensions are missing.
2. Were all the devices fabricated on the same substrate? Discuss the reproducibility and uniformity of such devices.
3. What advantages does dihydrated MAPbI₃ offer in this synaptic device compared with other perovskite or optoelectronic materials?
4. The authors report 20 operational cycles and 21 days of stability. How about long-term endurance, cycling, and retention analysis to clearly evaluate device lifetime and degradation mechanisms?
5. Since synaptic operations rely on illumination periods of tens of seconds and recovery times on the order of minutes, the responses are significantly slower than many reported optoelectronic synapses. A comparative discussion with state-of-the-art devices and possible strategies to improve switching speed would strengthen the manuscript.
6. The role of humidity in modulating synaptic plasticity is intriguing, but it raises concerns about practical deployment. Could the authors comment on encapsulation methods, environmental control strategies, or whether humidity dependence could be engineered into a functional feature rather than a limitation?
7. The methodology for processing data in simulating classification accuracies using RNN and D2NN is not fully clear.
8. In addition, the reported forgetting time (~5 s) is much shorter than in other optosynapses. Does this short retention influence the accuracy or reliability of the RNN and D2NN simulations?
9. How do physical variability, device-to-device fluctuations, and noise levels affect the performance of RNN and D2NN?

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

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 Chen,

Thank you once again for submitting your manuscript, "Multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonic synapses," to Communications Materials. It has now been seen again by the referees, whose comments are appended below. The concerns of our reviewers have now been addressed, but there are some amendments needed before we can accept your paper.

From Editor:

1. In particular, **you must use the following edited Abstract**, where inappropriate words have been removed, otherwise we are not able to publish your manuscript.

Optical neuromorphic computing utilizes light-based neural networks for high-speed, energy-efficient AI processing. Conventional optoelectronic synapses are limited by single-dimensional perception and electrical weight modulation. This study overcomes these constraints by developing all-photonic artificial synapses using water-mediated phase transitions in MAPbI₃ perovskite. These synapses exhibit excellent reversible light-driven optical memory capabilities, achieving a transmittance modulation range from 23.5% to 81.6% at 600 nm wavelength via precisely controlled crystal deformation mechanisms. The synaptic devices demonstrate high stability, maintaining consistent performance through 150 operational cycles and 6 months of environmental exposure. They successfully replicate essential neurobiological functions, including paired-pulse facilitation, short-term to long-term memory transition, and humidity-dependent plasticity. Implemented within a recurrent neural network, the system achieves 100% classification accuracy for multidimensional optical stimuli encompassing power, duration, and environmental humidity parameters. Furthermore, integration with a diffractive deep neural network enables reconfigurable computing with 80 distinct programmable transmittance states, achieving remarkable classification accuracies of 87% for MNIST handwritten digits and 76% for Fashion-MNIST datasets without requiring hardware modifications. This work establishes a paradigm for developing intelligent optoelectronic systems capable of adapting to complex environmental changes, demonstrating potential for applications in dynamic visual perception and multi-task processing environments.

From Assistant Editor:

1. Supplementary Information must be provided as a single file in PDF format.
2. Please specify if custom codes were developed or not during the study

We ask that you edit your manuscript according to the points above. **Please read this document carefully as we will be unable to further assess your revised paper until these important points are addressed.**

Please outline all revisions made in the author cover letter.

Please use the link below to submit your revised files:

Link Redacted

When resubmitting, please provide a marked-up manuscript with all changes highlighted, as well as a clean version of your paper.

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,

Sunkook Kim, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0003-1747-4539

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

My concerns are all solved now. I recommend the publication in its current form.

Reviewer #2 (Remarks to the Author):

The authors have addressed all my concerns. I recommend this paper for publication.

Reviewer #3 (Remarks to the Author):

The author's revisions have substantially improved the manuscript and have appropriately addressed the reviewers' comments. Therefore, I support the acceptance of this manuscript for publication in Communication Materials.

Version 2:

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 Chen,

Thank you once again for submitting your manuscript, "Multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonic synapses," to Communications Materials. The concerns of our reviewers have been addressed, but there are some amendments needed before we can accept your paper.

Please make the following changes to your letter:

- Please edit the abstract so that it does not exceed 160 words.
- An 'Introduction' heading is needed.
- The 'Results' heading should be replaced with 'Results and Discussion'

We ask that you edit your manuscript according to the attached table. **Please read this document carefully as we will be unable to further assess your revised paper until these important points are addressed.**

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.

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

When resubmitting, please provide a marked-up manuscript with all changes highlighted, as well as a clean version of your paper.

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,

Sunkook Kim, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0003-1747-4539

Version 3:

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 Chen,

Thank you once again for submitting your manuscript, "Multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonic synapses," to Communications Materials. Your manuscript has been seen again by the referees, whose comments are appended below. I am happy to say that the concerns of our reviewers have now been addressed, and that we only require some minor amendments before we can accept your paper.

Our remaining requests are:

- 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."

Figure files must only contain images (please leave out labels and titles such as "Figure 1" etc). Figure captions must instead be included within the main manuscript file, grouped together at the end of the document.

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.

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,
Sunkook Kim, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0003-1747-4539

Version 4:

Decision Letter:

Dear Professor Chen,

We are delighted to accept your manuscript titled "Multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonics synapses" 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](https://www.nature.com/commsenv/editorial-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](https://support.springernature.com/en/support/solutions/articles/6000281821-what-is-an-article-in-press).

Publication as an [Article in Press](https://support.springernature.com/en/support/solutions/articles/6000281821-what-is-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](https://forms.office.com/e/ed7NBDd08u), 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](https://www.nature.com/documents/Author_FAQs.pdf).

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.

If you have any questions about our publishing options, costs, Open Access requirements, or our legal forms, please contact ASJournals@springernature.com

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 <https://resource-cms.springernature.com/springer-cms/rest/v1/content/18943626/data/Research-Permission-Template-EN> Licence to Publish form. Please note that images should be supplied as 1400x400-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,
Sunkook Kim, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0003-1747-4539

*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 <http://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.

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 comments of the reviewer

Reviewer #1

The manuscript presents an intriguing approach to multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonics synapses. The work is timely and addresses an important direction in optical AI hardware. However, the conceptual basis of the synaptic weight definition and several technical details require substantial clarification. In particular, the equivalence of transmittance change (ΔT) with synaptic weight is not sufficiently justified and poses a fundamental concern. I therefore recommend major revision before further consideration.

1) Definition of Synaptic Weight (ΔT as weight):

The central issue is that the manuscript defines synaptic weight solely as the change in transmittance (ΔT). While ΔT may serve as an optical analogue to conductance modulation, its physical meaning, reproducibility, and comparability to established synaptic weights are insufficiently justified. This definition risks reducing the work to a simple demonstration of optical modulation rather than a robust synaptic weight implementation.

The authors should:

- Clarify how ΔT quantitatively maps onto weighted summation in optical neural networks.

Response: We sincerely thank the reviewer for raising this fundamental and critical

point regarding the definition of synaptic weight in our work. Actually, there might be a misunderstanding we need to clarify. The transmittance change (ΔT) is not used as the weight in our paper. In the RNN-based multidimensional classification (Fig. 4), the time-dependent ΔT is used as an input and processed by the RNN. In the reconfigurable D²NN-based classification (Fig. 5), the weights are derived from a programmable absolute transmittance T , which offers 80 distinct states. The multiply-accumulate (MAC) operation is achieved through the physical process of light-matter interaction and diffraction, as detailed below and consistent with established models for diffractive optical networks [*Lin, X. et al. All-optical machine learning using diffractive deep neural networks. Science 361, 1004–1008 (2018)*]. The transformation of an input optical field vector $I = [I_1, I_2, \dots, I_m]^T$ to an output vector $I' = [I'_1, I'_2, \dots, I'_n]^T$ through a layer of synapses is described by:

$$D \begin{pmatrix} T_1 \\ \vdots \\ T_i \end{pmatrix} = \begin{bmatrix} T_{1,1} & \cdots & T_{1,n} \\ \vdots & \ddots & \vdots \\ T_{m,1} & \cdots & T_{m,n} \end{bmatrix} \quad (1)$$

$$\begin{bmatrix} I'_1 \\ \vdots \\ I'_n \end{bmatrix} = \begin{bmatrix} T_{1,1} & \cdots & T_{1,n} \\ \vdots & \ddots & \vdots \\ T_{m,1} & \cdots & T_{m,n} \end{bmatrix} \cdot \begin{bmatrix} I_1 \\ \vdots \\ I_m \end{bmatrix} \quad (2)$$

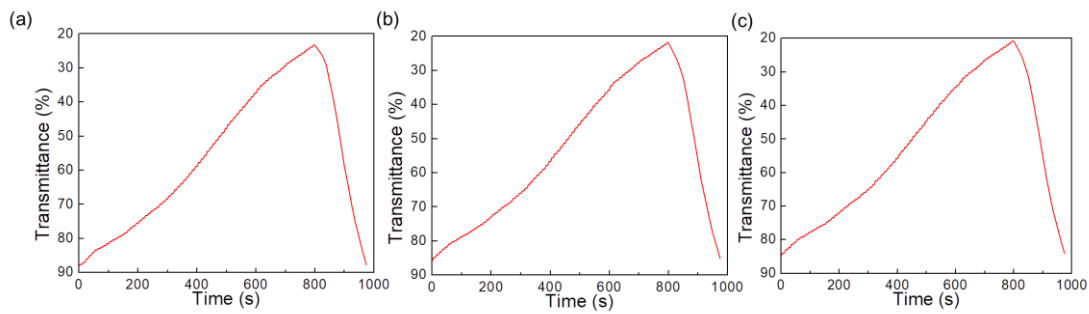
Where $T_1 \cdots T_i$ is the transmittance weight matrix (each element is one of 80 state values), D is the diffraction propagation function. This operation inherently performs the weighted summation in parallel. The multiplication $\mathbf{T}_{i,j} \mathbf{I}_j$ is achieved by modulating the input field's amplitude through the synapse, and the summation is

physically performed by the coherent superposition of light waves at the output plane.

We have added a theoretical explanation regarding the D²NN on Page 14.

- Discuss whether ΔT can provide stable, reproducible weight states under practical conditions.

Response: We sincerely thank the reviewer for the insightful question. We clarify that in the proposed D²NN architecture, the weights are derived from the absolute transmittance (T) value, selected from 80 discrete states, rather than the transmittance change (ΔT). The 80 programmable weight states demonstrated in our original manuscript (Fig. 5b) are not merely achievable in a single run but are highly reproducible. We have repeated the multi-state programming experiment multiple times, and the results show consistent and distinct transmittance levels (new data incorporated into Supplementary Fig. 19). The corresponding discussions have been added on Pages 14 and 15.



Supplementary Fig. 19 | a-c Reproducible transmittance tuning of the all-photonic synapses at the first (a), the second (b), and the third cycle (c).

- Benchmark ΔT -based weight against conductance-defined weights in memristive or transistor-based synapses.

Response: We appreciate the reviewer's comment. The advantages of the

transmittance-based weight are its native compatibility with optical computing. The weight multiplication is performed passively and inherently by the physical interaction of light with the programmed perovskite layer. This process requires no electrical power for the computation itself and occurs at the speed of light. Furthermore, a single diffractive layer leverages the natural parallelism of optics, executing these multiply-accumulate operations simultaneously as the light field propagates through it. Once programmed, the synaptic weights of transmittances are non-volatile and maintained without power consumption. We hope this clarification satisfactorily addresses your question.

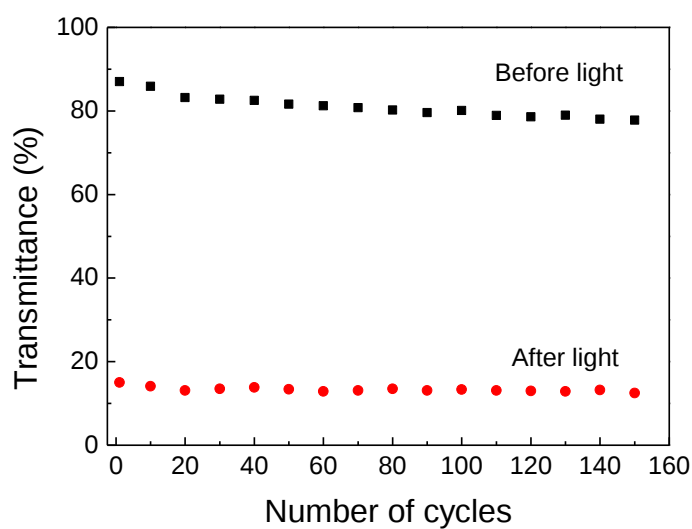
2) Stability and cycling data:

The manuscript provides stability tests (20 cycles, 21 days), which are relatively limited. Longer-term cycling or at least a discussion of retention, variability, and device-to-device reproducibility is needed.

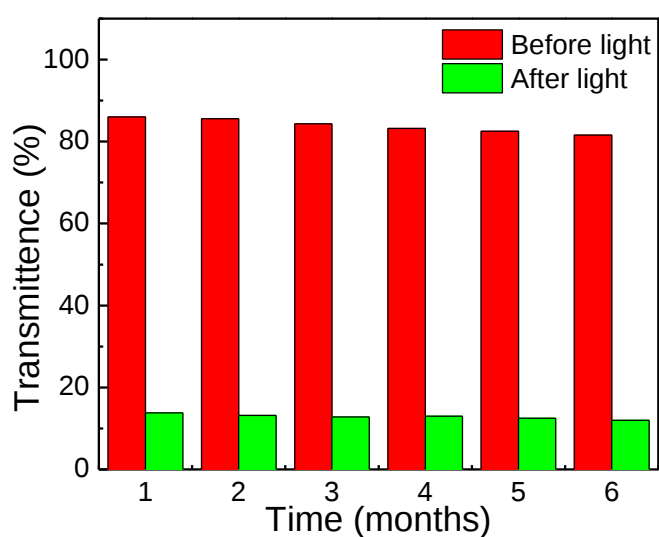
Response: We sincerely thank the reviewer for the valuable suggestion. We conducted additional, more rigorous stability tests. 150 consecutive switching cycles were performed under identical operational conditions. The device exhibited marginal degradation in transmittance modulation throughout this extended testing, confirming its excellent operational endurance for repeated use. These new data are presented in the revised Supplementary Fig. 2. We also monitored the optical states of the perovskite film over 6 months under ambient conditions. Both the hydrated (initial) and dehydrated (post-illumination) states retained their transmittance values without

noticeable decay. These results are now shown in the updated Supplementary Fig. 3.

These comprehensive results have been incorporated into the revised manuscript on Page 6.



Supplementary Fig. 2 | Cyclic performance test of a $(\text{MA})_4\text{PbI}_6 \cdot 2\text{H}_2\text{O}$ film.



Supplementary Fig. 3 | Stability of a $(\text{MA})_4\text{PbI}_6 \cdot 2\text{H}_2\text{O}$ film at ambient conditions under 50% RH. The film is illuminated under 383 mW with a duration of 60 s

To assess device-to-device reproducibility, we fabricated five all-photonic synapses

using the same synthesis protocol. The $\Delta T\%$ under the same stimulus was measured for each device. The results are summarized in the new Supplementary Table 2 (see below), which shows an average ΔT of 59.1% with a standard deviation of $\pm 2.2\%$. This narrow distribution demonstrates the high reproducibility of the synapse.

Supplementary Table 2: Reproducibility of the transmittance change across (MA)₄PbI₆·2H₂O films

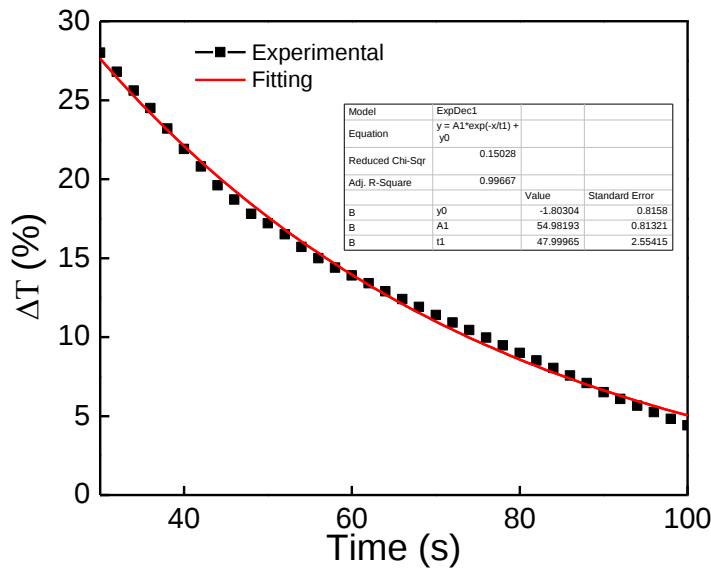
Film number	ΔT at 600 nm (%)
Film 1	60.5
Film 2	58.9
Film 3	61.2
Film 4	57.5
Film 5	57.4
Average $\pm$ Std	59.1 $\pm$ 2.2

3) Abrupt Optical Transitions:

The transmittance change in some plots appears abrupt, suggesting threshold-like behavior. A logarithmic scale or more detailed fitting would clarify whether the switching is analog or binary-like.

Response: We sincerely thank the reviewer for this insightful observation regarding the transition characteristics. We have re-plotted the key transmittance curves (revised Fig. 2c) with significantly higher data density, clearly revealing the continuous evolution of transmittance. Furthermore, we have performed a systematic kinetic analysis of the recovery process (Supplementary Fig. 7) of Fig. 3a. The trajectory at 60% RH fits well to a continuous exponential decay function, confirming that the switching mechanism is fundamentally analog and continuous. This exponential

relaxation behavior aligns with established models for continuous dynamics in synaptic devices. The corresponding revisions have been made on Page 9.



Supplementary Fig. 7 | Fitting result of transmittance change decay after 340 mW 365 nm illumination under 60% RH.

Reviewer #2 (Remarks to the Author):

The manuscript entitled "Multidimensional and reconfigurable optical neuromorphic computing using perovskite-based all-photonic synapses" written by Zi, Sun, and Chen et al. reports an all-photonic synapse based on photochromic dihydrated perovskites and demonstrates multidimensional as well as reconfigurable optical neuromorphic computing. The concept is interesting, and the experimental results clearly demonstrate synaptic plasticity and the high accuracy of RNN and ANN. However, there remain several points that require further clarification before the work can be considered for publication. The detailed questions are listed below.

1. Figure 3 shows that the device exhibits a markedly faster increase in transmittance during the re-learning process than during the initial learning cycle. Could the authors clarify the physical mechanism underlying this phenomenon?

Response: We sincerely thank the reviewer for this insightful question. This phenomenon is primarily attributed to the incomplete structural recovery of the perovskite film after the initial learning (dehydration) and subsequent partial forgetting (rehydration) cycle, which creates a metastable state that facilitates a faster response upon re-stimulation. The recovery during the forgetting cycles is insufficient for the film to fully revert to the pristine, well-ordered hydrated state. Instead, it remains in a metastable configuration with residual structural defects or pathways from the previous dehydration. This metastable state, being structurally closer to the dehydrated MAPbI_3 phase, presents a lower energy barrier for water egress upon re-illumination, thereby leading to the accelerated transmittance change observed during re-learning. The mechanism is supported by prior work on the reversible hydration-dehydration phase transitions in dihydrated perovskites [Halder et al., J. Phys. Chem. Lett. 2015, 6, 3180–3184]. Following the reviewer's suggestion, we have revised the manuscript on Pages 10 and 11 to include a clearer explanation of this mechanism.

2. Figure 4 presents classification tasks with relatively few classes and limited data, where one might expect even simple models to achieve high accuracy. While the RNN rapidly converges to 100%, the ANN does not. Could the authors elaborate on the factors that prevented the ANN from reaching perfect accuracy?

Response: We sincerely thank the reviewer for this insightful comment regarding the performance difference between the RNN and ANN models. The key reason the ANN failed to achieve perfect accuracy is the fundamentally temporal nature of our input data, which the ANN architecture is ill-suited to. However, the RNN is specifically designed to model such temporal sequences. It learns the characteristic shapes and evolution patterns of the transmittance responses, recognizing patterns such as a rapid increase followed by a slow decay, rather than relying on precise absolute values at specific time points. Our previous work on artificial olfactory synapses (Adv. Mater. Technol. 2024, 2301814) also reported an identical performance gap on the same classification task, where an RNN achieved >90% accuracy immediately, while an ANN struggled significantly. We have chosen to emphasize this architectural distinction on Pages 11 and 12 of the revised manuscript.

3. A conventional D²NN typically leverages different input modalities (e.g., distinct wavelengths or polarization states) to process and discriminate multiple datasets. However, in Figure 5, the presented "reconfigurable D²NN" seems to sequentially train and test the same network on two different datasets (MNIST and Fashion-MNIST) under identical optical conditions. From this perspective, the system appears to be more "reusable" than "reconfigurable." Could the authors clarify what distinguishes their approach as reconfigurable rather than merely sequential reuse, given that the same wavelength, humidity, and power conditions were maintained?

Response: We sincerely thank the reviewer for this insightful question. In our work, "reconfigurability" refers to the dynamic reprogramming of optical synaptic weights

for multiple tasks. Switching from MNIST to Fashion-MNIST classification requires loading a completely new set of weights, effectively creating a new neural network within the same physical hardware. This aligns with the concept of reconfigurable computing, where the hardware's function is altered post-fabrication through software-like control of its internal parameters. [Zhou, T. et al. Large-scale neuromorphic optoelectronic computing with a reconfigurable diffractive processing unit. *Nat. Photonics* **15**, 367–373 (2021)]. In our opinion, a "reusable" system implies using a static set of weights to process different datasets sequentially. Its weights are fixed, but can be "reused" for various inputs. We have added a related discussion on Pages 13 and 14.

4. If these all-photonic perovskite synapses are to be integrated into practical hardware, how would they operate within a neural network architecture? In electronic systems, input and output neurons are represented by voltage vectors; synaptic weights correspond to conductance values; and computation is governed by Kirchhoff's law. Similarly, how can all-photonic perovskite synapses be employed to perform neural network operations? A conceptual outline or framework for such an optical computing architecture would be highly valuable.

Response: We appreciate the reviewer's insightful question regarding the integration of all-photonic synapses into a functional computing architecture. The core computational framework, consistent with established optical computing models [Lin et al., *Science* **361**, 1004 (2018)]. Here, $\mathbf{I}$ is the input optical field vector, and $\mathbf{I}'$ is the output vector, D is the diffraction propagation function, $T_1 \cdots T_i$ is the transmittance

weight matrix (each element is one of 80 discrete values). The overall transformation is described by:

$$D \begin{pmatrix} T_1 \\ \vdots \\ T_i \end{pmatrix} = \begin{bmatrix} T_{1,1} & \cdots & T_{1,n} \\ \vdots & \ddots & \vdots \\ T_{m,1} & \cdots & T_{m,n} \end{bmatrix} \quad (1)$$

$$\begin{bmatrix} I'_1 \\ \vdots \\ I'_n \end{bmatrix} = \begin{bmatrix} T_{1,1} & \cdots & T_{1,n} \\ \vdots & \ddots & \vdots \\ T_{m,1} & \cdots & T_{m,n} \end{bmatrix} \cdot \begin{bmatrix} I_1 \\ \vdots \\ I_m \end{bmatrix} \quad (2)$$

This operation inherently performs the weighted summation in parallel. The multiplication T_{ij} is achieved by modulating the input field's amplitude through the synapse, and the summation is physically performed by the coherent superposition of light waves at the output plane. Following the reviewer's suggestion, we have added a discussion summarizing this conceptual framework on Page 14 of the manuscript.

Reviewer #3 (Remarks to the Author):

The authors have presented the development of perovskite-based all-photonic synapses for multidimensional and reconfigurable optical neuromorphic computing.

However, several key areas require further clarification and improvement:

1. The fabrication details of the synaptic device and its physical dimensions are missing.

Response: We sincerely thank the reviewer for raising this important point. The following details have been added to Page 17. The all-photonic synaptic devices were fabricated on glass substrates ($25 \times 25 \times 0.7$ mm). Before film deposition, the

substrates were ultrasonically cleaned sequentially in deionized water, anhydrous ethanol, acetone, and isopropanol, each for 15 minutes. Subsequently, the substrates were dried under a N₂ stream and treated with a UV-ozone cleaner for 25 minutes to enhance surface hydrophilicity. The perovskite precursor solution was then spin-coated onto the pre-treated substrates inside a glovebox, as described in the original manuscript.

2. Were all the devices fabricated on the same substrate? Discuss the reproducibility and uniformity of such devices.

Response: We sincerely thank the reviewer for raising this important point regarding the reproducibility and uniformity of our devices. All the devices characterized in this study were fabricated on the same type of glass substrates (25 × 25 × 0.7 mm) under identical processing conditions within the same batch. To comprehensively address the uniformity, we characterized both the physical thickness and the functional optoelectronic response across a single substrate. The film thickness, measured at five locations, averaged 495 ± 4.8 nm (**Supplementary Table 1**), confirming excellent physical uniformity. Moreover, the transmittance change (ΔT) showed high uniformity, with an average of 58.5% ± 0.5% under the same stimulus.

Supplementary Table 1 | Uniformity test of transmittance change and thickness on a (MA)₄PbI₆·2H₂O film

Measurement Position	Film thickness (nm)	ΔT at 600 nm (%)
1 (Center)	498	59.2
2	485	57.9
3	495	58.1

4	490	58.8
5	492	58.5
Average $\pm$ Std	492 ± 4.8	58.5 ± 0.5

To assess batch-to-batch reproducibility, we independently fabricated five perovskite films. The ΔT under the same stimulus was measured. As shown in Supplementary Table 2, an average ΔT of 59.1% with a standard deviation of $\pm 2.2\%$ is measured, demonstrating the high reproducibility of the proposed fabrication process. The corresponding data and discussion have been added on Page 5.

Supplementary Table 2 | Reproducibility of the transmittance change across (MA)₄PbI₆·2H₂O films

Film number	ΔT at 600 nm (%)
Film 1	60.5
Film 2	58.9
Film 3	61.2
Film 4	57.5
Film 5	57.4
Average $\pm$ Std	59.1 ± 2.2

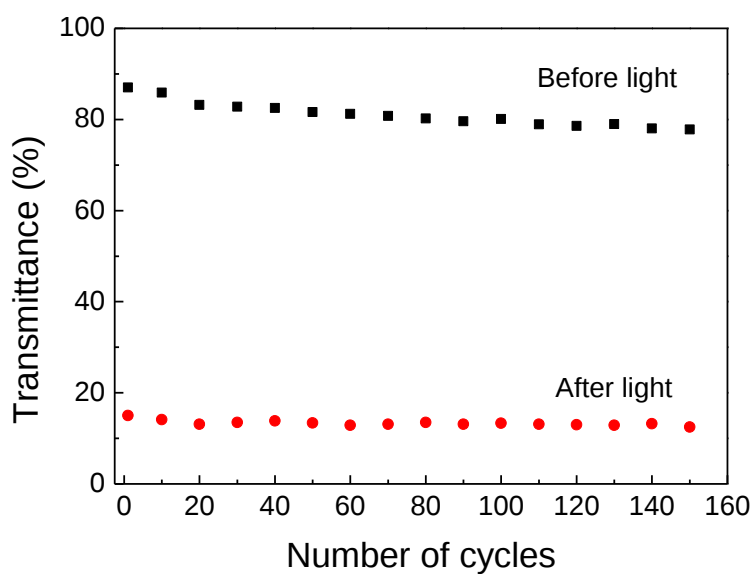
3. What advantages does dihydrated MAPbI₃ offer in this synaptic device compared with other perovskite or optoelectronic materials?

Response: Thanks for this question. Dihydrated MAPbI₃ offers several advantages. First, its response relies on light-induced transmittance modulation, eliminating electrical wiring and enabling circuit-free, parallel optical computing at high speed and with high energy efficiency. Second, the crystal transformation enables large

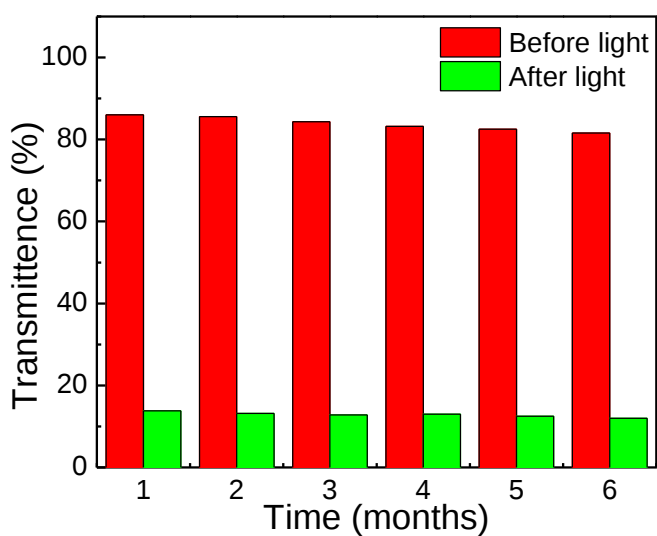
transmittance modulation (up to 66%), offering a broad dynamic range for synaptic weights. Third, ambient humidity natively modulates the phase transition, serving as an intrinsic control parameter for synaptic plasticity and multidimensional signal processing. Finally, the perovskite transmittance can be optically maintained under low humidity and tuned by light illumination, providing a pathway to achieve multi-task neuromorphic computing without hardware replacement.

4. The authors report 20 operational cycles and 21 days of stability. How about long-term endurance, cycling, and retention analysis to clearly evaluate device lifetime and degradation mechanisms?

Response: We thank the reviewer for highlighting the importance of long-term device stability. We have conducted extended endurance and retention tests. The device maintains its transmittance modulation without significant degradation over 150 cycles (Supplementary Fig. 2), demonstrating excellent operational endurance. Furthermore, both the hydrated and dehydrated states show marginal transmittance decay after 6 months under ambient conditions (Supplementary Fig. 3), confirming exceptional long-term retention and environmental stability. The corresponding discussion has been added on Page 6.



Supplementary Fig. 2 | Cyclic performance test of a $(\text{MA})_4\text{PbI}_6 \cdot 2\text{H}_2\text{O}$ film.



Supplementary Fig. 3 | Stability of a $(\text{MA})_4\text{PbI}_6 \cdot 2\text{H}_2\text{O}$ film at ambient conditions under 50% RH. The film is illuminated under 383 mW with a duration of 60 s.

5. Since synaptic operations rely on illumination periods of tens of seconds and recovery times on the order of minutes, the responses are significantly slower than

many reported optoelectronic synapses. A comparative discussion with state-of-the-art devices and possible strategies to improve switching speed would strengthen the manuscript.

Response: We thank the reviewer for raising this important point regarding the device speed. We acknowledge that the phase transition in the dihydrated MAPbI₃ film, driven by hydration/dehydration dynamics, results in slower switching compared to optoelectronic synapses. We are actively exploring material-level optimizations, such as elemental doping, nanostructuring, and crystallinity control, to accelerate the ion migration and phase transition in future work. On the other hand, it is crucial to distinguish the response time of the material from the computational speed of the neural network. The computation in the D²NN is executed at the speed of light once the weights are set, which is much faster than optoelectronic synapses. In this case, the proposed computing with simplicity, high processing speed, and excellent environmental adaptability is particularly suited for applications where ultrafast weight updates are secondary to high processing speed, including edge AI, adaptive optical preprocessing, robust vision systems, and reconfigurable holography. A corresponding discussion has been added on Page 16.

6. The role of humidity in modulating synaptic plasticity is intriguing, but it raises concerns about practical deployment. Could the authors comment on encapsulation methods, environmental control strategies, or whether humidity dependence could be engineered into a functional feature rather than a limitation?

Response: We sincerely thank the reviewer for this valuable comment. Rather than viewing this solely as a limitation, an encapsulation strategy that transforms humidity sensitivity into a programmable feature can be proposed. The strategy involves locating the device in a sealed transparent chamber equipped with a gas-permeable interface and a sealable port. This design enables two key operational modes. In a standard operating environment, the chamber can be sealed to lock in a stable humidity level, ensuring consistent device performance. When humidification is desired, the port can be opened to enable precise humidity control with a micro-humidifier. The strategy effectively engineers humidity dependence into a functional advantage, enabling the creation of adaptive optical systems that can be erased and reconfigured.

7. The methodology for processing data in simulating classification accuracies using RNN and D²NN is not fully clear.

Response: We sincerely thank the reviewer for this valuable comment. We have now substantially revised the manuscript to provide a comprehensive description of the RNN and D²NN data processing methodology. These revisions are located in the Methods section under "Neuromorphic Computing" on Pages 18 and 19.

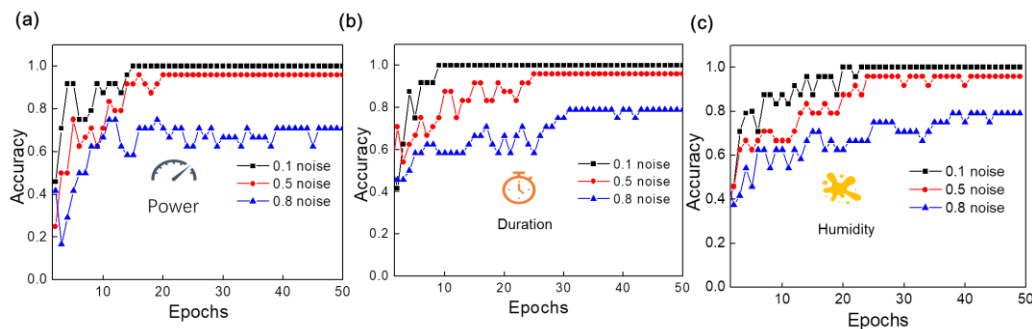
8. In addition, the reported forgetting time (~5 s) is much shorter than in other optosynapses. Does this short retention influence the accuracy or reliability of the RNN and D²NN simulations?

Response: We thank the reviewer for raising this important point. Actually, the short forgetting time under specific conditions does not affect the accuracy of RNN- and

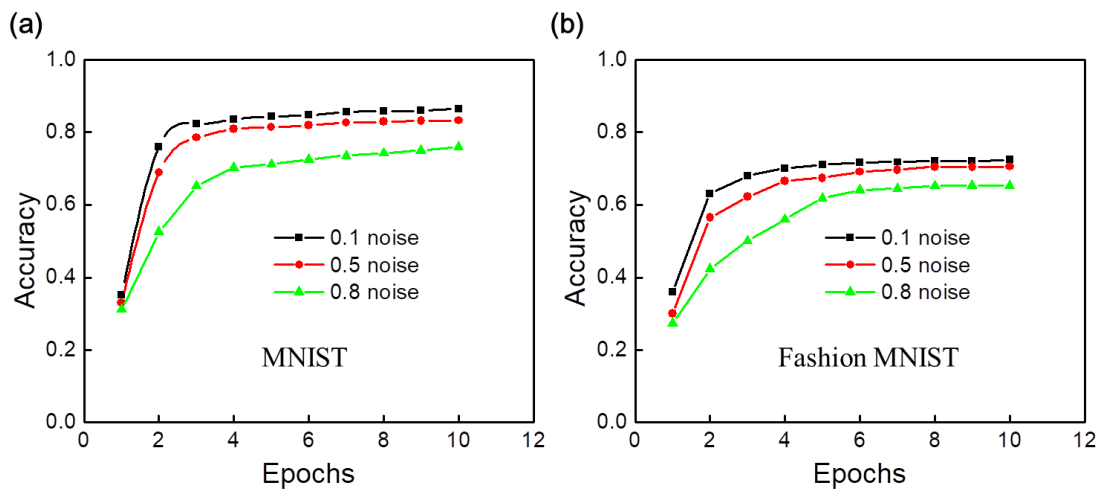
D²NN-based classifications. For RNN-based classification, accuracy depends on analyzing the temporal features of the transmittance response rather than on prolonged state retention. For the D²NN-based classification, all programming and operations are conducted under stable, low-humidity conditions (30%), where the synaptic weights exhibit excellent non-volatile retention. As shown in Supplementary Fig. 19, the programmed weights remain stable, ensuring the reliability and accuracy of the optical inference.

9. How do physical variability, device-to-device fluctuations, and noise levels affect the performance of RNN and D²NN?

Response: We sincerely thank the reviewer for raising this important question regarding the impact of physical variability and noise on system performance. We have conducted comprehensive robustness analyses for both our RNN and D²NN systems. For the RNN-based classification tasks, we introduced additive Gaussian noise with standard deviations of 0.1, 0.5, and 0.8 to the input signals. The RNN demonstrated strong robustness, maintaining over 95% accuracy for power, duration, and humidity recognition at noise levels up to $\sigma = 0.5$. Even under extreme noise ($\sigma = 0.8$), the RNN retained accuracies above 71–79% (Supplementary Fig. 9). We have added a detailed discussion of these findings on Page 12.



Supplementary Fig. 9 | a-c Power (a), duration (b), and humidity (c) recognition accuracies processed by a RNN with different standard deviations of Gaussian noises. For the D²NN, similar noise analysis revealed remarkable resilience, at low noise ($\sigma = 0.1$), classification accuracy on MNIST remained near 86%, matching the noise-free performance. Even at high noise levels ($\sigma = 0.8$), the system retained over 75% accuracy as shown in Supplementary Fig. 24. This robustness stems from the inherent parallelism of optical diffraction and noise-invariant feature learning during training. We have added these new results and corresponding discussion to the revised manuscript (Page 16).



Supplementary Fig. 24 | MNIST (a) and Fashion-MNIST (b) recognition accuracies processed by a D²NN with different standard deviations of Gaussian noises.