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Halide perovskite quantum dots photosensitized-amorphous oxide transistors for multimodal synapses

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Abstract:

Deployment of novel artificial synapses serves as the crucial unit for building neuromorphic hardware to drive data-intensive applications. Emulation of complex neural behaviour through conventional Si-based devices requires a large number of elements which increases fabrication complexity and brings challenges of connectivity. Hence, there is a need to investigate alternative material systems and device architectures for emulating richer neural behaviour comprising of lesser elements. Herein, a thin-film transistor (TFT)-like synaptic device using all-inorganic Cesium lead bromide (CsPbBr₃) perovskite quantum dots (QDs) and amorphous Indium Gallium Zinc Oxide (a-IGZO) semiconductor active material is explored for brain-inspired computing. The incorporation of CsPbBr₃ QDs as a photosensitizer aids in realizing light-dependent synaptic memory. Furthermore, type II heterostructure can serve as a basis for electro-optical programming. The proposed artificial synapse demonstrates a materials combination that could decouple optical absorption and charge transport property, provides freedom to tune the spectral region. Harnessing the advantages of novel materials, our devices obey spike-timing-dependent plasticity rules, inculcate associative learning and linear non-volatile blind updates. This architecture paves way for efficient building of neuromorphic hardware elements with facile tunability and tailorable plasticity.

Keywords: perovskite quantum dots photosensitizer, oxide semiconductor, interfacial charge transfer, artificial synapses, non-volatile memory

Introduction:

Traditional computing based on von-Neumann architecture or Boolean logic are well-suited for solving systematic problems, such as predefined mathematical models or resolving structured datasets. However, they are unable to match the human brain that can solve higher level challenges by interacting with the real world even when inputs and outputs are imprecisely specified. Neuromorphic computing- an approach to build electronic devices and circuits analogous to neurons and synapses in the human brain, promises to dramatically improve the performance of big data tasks efficiently^[1]. The main difference between neuromorphic computing and von-Neumann architecture-based computing are in the way the information is being processed, i.e., integrated memory and processing in neuromorphic computing vs. separate memory and central processing unit (CPU) in conventional computers^[2,3]. In non-von Neumann computing architecture based on human brain, neurons are considered as processors and synapses are considered as memory elements. The separated memory-processing in von-Neumann architecture causes a limitation in the data throughput which is exacerbated for big data applications (also known as von Neumann bottleneck)^[2,4]. Therefore, such an in-memory computational architecture is essential to solve the bottleneck issues. The human brain is adaptive in task handling that involves real-time sensory signals such as touch, image, and sound with ultralow power consumption (~ 20 W) ^[5]. The attributes of the human brain such as learning, parallel processing^[6], fault-tolerance and integrated memory and processing seems unachievable in conventional von-Neumann architecture^[7,8].

Emulation of artificial neural networks in software level is widely studied, however, hardware-level execution is still limited owing to its dependency on traditional CMOS technology. Despite the remarkable progress of nanoscale two-terminal memristors, the abrupt switching physics of most memristive solutions often limits the number of addressable states to conventional binary logic, limiting their plasticity and storage capacity that adversely affects the trainability of artificial neural networks (ANNs)^[9]. Efficient training of such ANNs hinges on the incremental adjustment of weighted connections between the hidden layers, in turn demanding deployment of multiple programmable conductance states ^[3,4]. With the scaling limits continuously challenged by the inherent complexity of manufacturing and photolithography, strategies to elevate the storage density of artificial synapses in ANNs necessitate the development of neuromorphic systems comprised of lesser elements displaying richer neuromorphic phenomenon^[10]. Since conventional memristors only possess two terminals, the readout and programming occurs through the same set of terminals, which could

be a limiting factor^[11,12]. Conversely, three-terminal devices have more control with the gate electrode used for programming and the source-drain terminals used for read-out^[3,6,13]. Ideally, a three-terminal transistor system where additional control inputs could effectively ‘gate’ the memconductance of the active switching element, would enable augmented conductance/weight modulations that standard two-terminal memristive implementations fail to address^[14,15]. In addition, an “optical” mode of writing/accessing of memory states, independent of “electrical” programming schemes, would increase the storage capacity^[16] of such transistors utilizing the higher bandwidth associated with optical signals^[17,18]. Moreover, physical interconnections could be substituted thereby reducing thermal loss at the system level^[19]. In this respect, novel materials that could demonstrate the functionalities of optoelectronic synapses are still in the early stages of development^[18,20]. The choice of the active electronic material simultaneously modulates the optical and electrical properties which could be a limiting factor. A materials combination that could decouple optical absorption and charge transport would allow for facile tunability^[21]. Having an independent choice of optical absorber provides the freedom to tune the spectral response^[22]. Such tunable synapses would result in the activation of specific synapses but not others. For example, blue light-sensitive synapses would respond to optical wavelengths ($\lambda > 445$ nm) while not responding to red light ($\lambda = 620$ nm). This kind of multiplexing can be of greater use for selectivity and control in learning^[23]. Photo-responsive materials offer a huge advantage for constructing artificial synaptic devices. It aids in converting incident light signals into volatile or non-volatile electrical signals. The integrated architecture of the synaptic device with light-sensitive elements mimics the visual system by converting visible light photons into synaptic signals. Rods and cones from the human eye interact with the incident light and transmit signals through synapses to retina bipolar cells. Inspired by biological visual perception, integrating photodetectors with memory element aids in achieving visual non-volatile memory. To build an efficient artificial cogni-retina device for the robotic eye, there are two main prerequisites. i) a synaptic element that captures visual data which is sensitive to both photonic and electronic stimulations to adjust its synaptic states (LTP and LTD) ii) able to mimic basic functions associated with the visual cortex.

Here we harness the electrical and optical perturbation of charge carriers and their trapping-detraping dynamics across the novel perovskite quantum dots (cesium lead bromide-CsPbBr₃) - amorphous oxide semiconductor (Indium Gallium Zinc Oxide-IGZO) heterojunctions to design thin-film transistor (TFT) synaptic devices for brain-inspired neuromorphic computing.

Incorporating a photosensitizer (CsPbBr_3) on to the existing oxide semiconductor (a-IGZO) TFT aids in realizing light-dependent non-volatile memory states via optical inputs. All inorganic CsPbBr_3 QDs are an emerging class of halide perovskite material with its size-dependent optical properties^[24], stability, bandgap tunable^[25], display defect tolerance, and shows excellent absorption^[7]. The unique ionic-electronic charge transport property of CsPbBr_3 could be dynamically tuned to meet the demands of the analog switching in ANNs. Controlled injection and recombination of charge carriers across the type-II interface between CsPbBr_3 quantum dots and a-IGZO is demonstrated by the proper choice of material selection. The CsPbBr_3 -a-IGZO optoelectronic transistor responds to 445 and 525 nm wavelengths and allowing optoelectronic near-linear programming, necessary as an accelerator for deep learning in artificial neural networks (ANNs). Moreover, the photoplasticity modulated by CsPbBr_3 -a-IGZO heterostructure demonstrating adaptive optical perception under 445 and 525 nm, this artificial optoelectronic synapse could be feasible for mimicking human visual memory. The ability to modulate synaptic plasticity is more significant for the “optical” programming scheme when compared to its “electrical” counterpart. Optimized pulsing schemes with synergistic electro-optical modes aids in modulating short- and long-term memory, basis for learning and computing^[26,27]. To satisfy the programming/erasing requirements of ANNs, we utilized synergistic optical and electrical signals in our artificial synaptic TFT for inducing long-term conductance change. Here, optical pulsing schemes aids in achieving non-volatile writing, while electrical pulsing schemes aids in erasing weight states via electrical gating. Increase in conductance during “writing” operation correlates to the photocarriers in the semiconductor channel while decrease in conductance during “erasing” operation correlates in accelerating the recombination process, denotes a non-volatile write/erase memory cycle. Conditioned as a neuromorphic memory, our artificial synapse based on all-inorganic CsPbBr_3 -a-IGZO TFT demonstrate reconfigurability, associative learning and forgetting analogous to the human brain. The proposed IGZO synaptic transistor can act as a fundamental device with a variety of photosensitizers such as dyes, organic semiconductors, 2D materials, and chalcogenide quantum dots. Our methodology opens a novel and efficient way of building neuromorphic hardware elements with modal programming schemes and tailorable plasticity.

Results and discussion:

Figure 1. (A) Schematic of a biological synapse (left) and (B) the proposed multi-gated transistor as artificial synapses (right). (C) Cross-sectional FE-SEM image of the device architecture-IGZO/ITO/CsPbBr₃/PMMA. (D) Transmission electron microscopy (TEM) image and (E) High-resolution TEM image of CsPbBr₃ QDs (F) Transfer and (G) Output characteristics of the bilayer IGZO-CsPbBr₃ channel TFT in dark and light (blue “λ” - 445 nm, intensity – 8 mW/cm²) conditions. (H) Steady-state photoluminescence (PL) spectra of CsPbBr₃+PMMA and IGZO+CsPbBr₃+PMMA heterojunction. CsPbBr₃+PMMA displays a sharp emission at ~525nm with 400nm excitation wavelength. In contrast, the heterojunction of IGZO+CsPbBr₃+PMMA emits a much weaker signal, indicating effective charge transfer at the heterojunction.

Figures:1A-B illustrates the working principle of biological synapses and the analogy to our optoelectronic artificial synapses. The human brain contains a large number of interconnected neurons (10^{11}) and synapses (10^{15}) where signal transmission occurs^[28,29]. During an action potential, pre-synaptic neuron transmits the signal to the post-synaptic neuron in a junction called synapse. For example, when triggered by an action potential, the voltage-dependent Ca^{2+}

ions flow into a presynaptic neuron and move through distinct channels that opens in response to the changed voltage^[30]. These Ca^{2+} ions binds with the synaptic vesicle, triggering the vesicles to break and discharge neurotransmitters into the synaptic cleft as shown in Figure 1A. Subsequently, these neurotransmitters interact with binding sites on the post-synaptic neurons to commence the opening of ion-channels and depolarize or hyperpolarize the membrane potentials. The neurotransmitters then produce excitatory post-synaptic potential (EPSC) or inhibitory postsynaptic potential (IPSC) in post-synaptic neurons^[31]. Depending on the strength of the neural activity, the connection strength either strengthens (potentiation) or weakens (depression), and is termed as synaptic plasticity^[32]. i.e., the connection strength gets stronger for the active synapse and weaker for the inactive synapse.

For the multi-terminal memristive synapses (Figure 1B), the gate terminal serves as the presynaptic input while the source/drain electrodes act as the post-synaptic output terminals^[33,34]. Controlled injection and recombination of charge carriers across the heterojunction and semiconductor-dielectric interface mimic the role of ions and neurotransmitters in biology, while channel conductance serves as the synaptic weight. Figure:1C shows the cross-sectional field-emission scanning electron microscopy (FE-SEM) image of the proposed device architecture. A type-II heterostructure is formed by spin coating CsPbBr₃ QDs (~60nm) on sputtered IGZO (~60nm) thin films. ITO (~130nm) serves as the source and drain electrodes, while 15 nm of polymethyl methacrylate (PMMA) serves as the passivation. The morphological image of spin coated CsPbBr₃ QDs is shown in figure S1 of supporting information. Figure:1D shows the transmission emission microscopy (TEM) image of CsPbBr₃ QDs with an average diameter of ~10nm. The detailed structural property is studied using high-resolution (HR) TEM as shown in figure 1E. The lattice spacing of 0.59 and 0.48 is attributed to the <100> and <110> planes of the cubic phase CsPbBr₃ QDs. Small difference exists in the size and shape of the CsPbBr₃ QDs due to the size distribution among the nanocrystals (NCs). The average size of NCs are 9.6 ± 0.5 nm is evident by the histogram plot as explained in figure S2 of the supporting information. Tapping mode atomic force microscopy (NX 10) is used to study the morphological imaging of CsPbBr₃ QDs nanocubes. The average roughness of the spin-coated CsPbBr₃ QDs layer is 18.69 nm for 10 X10 μm^2 scanning area. The surface morphology and the corresponding 3D profile of the CsPbBr₃ QDs layer are shown in figure S3 of supporting information. Thin-film XRD (Bruker D8 Advance) is deployed to analyze diffraction peaks of CsPbBr₃ QDs. CsPbBr₃ QDs are synthesized through hot-injection method. A zero-diffraction substrate holder is used to reduce background noise. As observed from Figure S4-A, the obtained peaks of as-synthesized CsPbBr₃ quantum

dots matched with the cubic phase patterns in reported literature (ICSD – 04-005-6613)^[35,36]. Detailed synthesis and fabrication procedures are explained in the supporting information (SI-Note-1).

Figures 1F-G shows the transfer and output characteristics of the hybrid IGZO-CsPbBr₃ phototransistor under dark and light illumination. The devices exhibit a typical n-type enhancement-mode operation with field-effect mobility and on-off ratio of 5.07 cm² V⁻¹ s⁻¹ and 10⁶ respectively under dark conditions. Upon illumination, the devices exhibit a depletion-mode operation indicated by the increase in drain current and left shift of the threshold voltage (1V to -5V) for channel formation, suggesting the presence of strong photo-gating effects across the heterojunction. Additionally, a distinct clockwise hysteresis window of 1V is observed, indicates trapping and de-trapping of electrons, defining the memory window. On applying a small positive/negative electrical pulse at the gate (pre-synaptic) terminal, the channel conductance (weight) increased/decreased temporarily and quickly ceased to its initial conductance state or resting current. This temporary change in the channel conductance is related to the gate capacitance effects and the hysteresis observed is expected to arise from trap centres at the IGZO-SiO₂ interface. Larger magnitude of negative pulses causes a permanent decrease of channel conductance probably due to the residual carrier concentration in the IGZO layer in its off state. Figure 1H inset shows the energy band diagram of CsPbBr₃-IGZO and illustrates the working mechanism for optical programming. If the incident photon energy is greater than the bandgap of CsPbBr₃ QDs ($E_G > 2.4$ eV, $\lambda < 530$ nm), electron-hole pairs are generated. These photogenerated electrons are injected into IGZO generating photocurrent. The process results in increasing the channel conductance and left shift of threshold voltage (V_{th}) for channel formation. However, the photogenerated holes are trapped in CsPbBr₃ since the valence band of CsPbBr₃ is lower than the valence band of a-IGZO. These trapped holes act as a positively biased gate (photogating effect)^[37,38] that results in the increase of channel conductance. After light illumination is removed, activation energy is required to neutralize these trapped charges, thus the rate of recombination is slow. Subsequently, channel conductivity decays slower owing to the delayed removal of trapped charges. This phenomenon of delayed recombination and slow decaying current is called the Persistent Photoconductivity (PPC)^[39]. Figure S4-B shows the absorbance and photoluminescence spectra of CsPbBr₃ QDs (nanocubes). The as-synthesized CsPbBr₃ QDs shows strong optical absorption at 530 nm. As shown in figure S4 C-D, the absorption edge of IGZO and IGZO-CsPbBr₃ QDs is observed to be around 400nm and 550nm, indicating bandgaps of 3eV and 2.4eV respectively, matching well with previous reports^[40]. Furthermore, the charge transfer at

the heterojunction has been studied using UPS spectra as explained in figure S5 and S6 of supporting information. Configured as a “gated” memristive artificial synapse, we utilize these short-term hysteresis effects and long-term V_{th} shifts of the thin film transistor upon electrical and optical gating as a proxy for synaptic plasticity.

In neuromorphic architectures, artificial synapses act as weighted connections between layers of the neural network enabling energy-efficient in-memory computations^[41]. Hence to qualify as weighted synaptic connections in a neural network, the devices are first benchmarked on the degree of plasticity as a function of independent input electrical and optical stimuli, followed by synergistic electro-optical training schemes. Therefore, our synaptic devices that can retain memory states up to ~10s is defined as short-term memory and above 500s as long-term memory.

Electrical programming of synaptic weights: Categorised based on the operational timeline, short-term and long-term plasticity rules outlining learning and memory are realized by recording post synaptic currents (PSCs) I_{DS} as a function of the electrical pre- (V_G) and post-synaptic (V_D) voltage parameters like pulse interval, width, frequency and number of repetitions. Analogous to biological neural or paired-pulse facilitation (PPF), a pair of excitatory pulses (+5V, pulse width=0.5s, pulse interval=0.5s) at the pre-synaptic terminal evokes a paired pulsed current response in our artificial synapses, which immediately dropped back to its resting current of ~7nA when removing the input spike pattern (SI-Note-2 Figures:S7-C)^[33]. When the pulse interval between two presynaptic input spike pattern is kept <0.5s, the PPF index is noticed to be higher than 100%, demonstrating strong facilitation at such timelines, similar to the residual Ca^{2+} ion hypothesis by Katz and Miledi^[42]. For the shortest pulse interval of 0.5 s, our artificial synapses depicted the highest PPF index of 140%, followed by 134.5% and 129% at intervals 1s and 1.5s respectively. This ratio continues to decrease with increasing pulse interval and reached ~122% finally for the pulse interval of 3s (Figure:2A). In congruence with biologically observed values, the decay function could be fitted well with an exponential equation.

$$y = y_0 + B_1 * \exp\left(-\frac{x - x_0}{t_1}\right)$$

where x is pulse interval, y_0 is magnitude, B_1 is facilitation constant, and t_1 is time constant as shown in SI-Note-2 Table:T1. Such rules on short-term memory and plasticity have been shown to serve as working memory and mapping matrices to remove auto-correlations and have been adopted very lately to learn good temporal composition of event-based signals in the midst of rate-coded occurrences^[43]. The resting currents for short-term plasticity ranges

from 2-7 nA, as observed from figure 2c and S7 (a, c) with negligible conductance difference.

Figure 2. Electrical programming of synaptic weights. PPF indices as a function of the (A) inter-pulse interval and (B) pulse width respectively. Current transients depicting (C) short-term potentiation and (D) long-term depression, respectively. (E) Conductance map as a function of blind programming pulses. The updates are volatile in the potentiation phase and non-volatile in the depression phase (F) zoomed-in view of the same

Increasing the pulse width of the presynaptic voltage results in a controlled modulation of the PPF indices as shown in Figure:2B, indicating the modulatable nature of short-term plasticity in our devices. However, a pair of inhibitory pulses (-2 V, pulse width=500ms, pulse interval=500ms) at the pre-synaptic terminal evokes inhibitory non-volatile responses in our artificial synapses as shown in SI-Note-2 Figure:S7-D. On training with higher number of presynaptic pulses, volatile response or short-term potentiation remains consistent in the excitatory phase as shown in Figure:2C, SI-Note-2 Figure:S8-A. During electrical potentiation, when a positive voltage is applied at the gate terminal, the electric field drives holes towards the IGZO-CsPbBr₃ interface. Under the assistance of the electric field, the probability of hole trapping in the perovskite quantum dots/interfaces is high. Upon removal of the electric field,

positive gating due to the trapped holes increases electron accumulation in the channel. As a result, channel conductance increases (potentiation). However, we believe that these holes occupy shallow trap states and thermal energy is enough to result in the detrapping of the holes. This results in volatility under electrical potentiation conditions (supporting information figure S9 A). Similarly, a non-volatile response or long-term depression is consistent in the inhibitory phase as shown in Figure:2D, SI-Note-2 Figure:S8-B. During electrical depression, when a negative voltage is applied at the gate terminal, majority carrier electrons are driven and trapped at the IGZO-CsPbBr₃ interface. Driven by the gate voltages and the number of carriers it induces, the electrons can occupy deeper trap states. It is suggested that the activation energy (E_a) required to remove the trapped electrons from CsPbBr₃ could be relatively high. Since electron traps are deeper and spatially separated from holes, they have prolonged recombination lifetime. This mechanism explains the non-volatility of electrical depression under negative gate bias. The lower magnitudes of electrical potentiation as compared to electrical depression again point to minority carrier trapping during potentiation and majority carrier trapping during depression (supporting information figure S9 B). Despite the fact that stable long-term depression has achieved (electrical erasing), the inability to update conductance/weights in a non-volatile manner in the potentiation phase indicates the shortcoming of such devices to run spike-based learning rules such as Spike-Timing-Dependent Plasticity (STDP) in the pure electrical-mode. A plot of the current transients with blind programming pulses revealed small volatile and large non-volatile transitions in the potentiation and depression phases respectively (Figure:2E), resulting in a highly non-linear asymmetric response map, undesirable for ANNs. Hence, to probe non-volatile weight updates in the potentiation phase, optical programming schemes are employed in our devices as explained below.

Optical programming of synaptic weights: The excellent optical absorption and superior electronic properties of CsPbBr₃^[44] QDs allows for a wide modulation of the photo-generated carriers at the type-II heterojunction, in turn unlocking the possibility of addressing synaptic weights with optical pulses, mutually exclusive to the afore-described electrical programming schemes.

In congruence with Atkinson-Shiffrin's memory model^[45] (Figure:3A), our devices depicted stronger consolidation of long-term memory upon repeated and intense input optical stimuli and weakened (forgetting) with long intervals between rehearsals (Figures:3B-E)^[45]. Upon

excitation with blue and green light pulses (Blue “ λ ” = 445 nm, Green “ λ ” = 525 nm intensity = 12mW/cm²), a strong non-volatile response is induced in our devices as shown in Figure:3B. Shortest wavelength blue light (“ λ ”= 445 nm) illumination induces large postsynaptic current (I_{DS} = 280 nA), whereas green light (“ λ ”= 525 nm) illumination induce weak post synaptic current (I_{DS} = 235 nA) which corresponds to the strong absorption of CsPbBr₃ QDs in the ultraviolet region. The output response decays to a metastable state in >500s (defined here as long-term memory) as supposed to the short-term output responses which decayed back to its initial conductance state in <10s. For example, on increasing the light stimulus intensity from 8 to 16mW/cm², the degree of long-term potentiation (LTP) increases from 26% to 97% (Figure:3C). Similarly, the degree of LTP increases from 40% to 125% on increasing the pulse width from 1s to 2.5s (Figure:3D). In accordance with Ebbinghaus’s learning rules, repeated rehearsals result in stronger consolidation of long-term memory and decreased forgetting rates (Figure:3E).

In summary, electrical perturbation results in short-term potentiation (STP) and long-term depression (LTD), while optical perturbation results in long-term potentiation (LTP). This motivates us to utilize a combination of both electrical and optical programming pulses to suffice all the weight update requirements of ANNs. Hence, in the following section, we exploit synergistic multi-modal electro-optical programming schemes to benchmark the artificial synapses against the common learning rules of spiking [SNNs] and deep neural networks [DNNs].

Figure 3. Optical programming of synaptic weights. (A) The multistore modal memory model depicting short-term and long-term memory proposed by Atkinson and Shiffrin. Current transients depicting long-term potentiation upon optical stimulation. Function of (B) light wavelength, and (C) optical illumination intensity, (D) pulse width of illumination and (E) number of training pulses using blue light ($\lambda = 445$ nm).

Electro-Optical programming of synaptic weights facilitates Spike-Timing Dependent Plasticity rules, Linear Non-Volatile Blind Updates and Associative Learning:

Spike-Timing Dependent Plasticity (STDP): Synergistic electro-optical programming of synaptic weights facilitates to demonstrate spike-timing dependent plasticity (STDP) learning rules in our CsPbBr₃-a-IGZO systems. Depending on the timing between the pre-synaptic to postsynaptic neuron, voltage and time-based synaptic weight alters the conductance of the active material^[46]. As explained by Hebb's theory, various forms of STDP learning rules are realized in our system, since the dependency between the occurrence of pre-synaptic and post-synaptic events is crucial for learning and memory^[47]. If the post-synaptic input spikes are followed by pre-synaptic action potential, due to coordination between the pre- and post-synaptic action-potential in a short interval of time ($t_{\text{post-pre}} > 0$), the synaptic connection

strengthens resulting in long-term potentiation (LTP). Similarly, if the pre-synaptic input spikes are followed by post-synaptic action potential in a short interval of time ($t_{\text{post-pre}} < 0$), due to incoordination between the pre- and post-synaptic action-potential, the synaptic connection weakens resulting in long term depression (LTD). Here, optical-pre and electrical-postsynaptic input spikes are applied to the gate and drain terminals of the transistors by keeping source floated. The post-synaptic weight change (ΔW) is measured from the drain terminal. The effective weight changes due to the temporal correlated pre-synaptic input spikes lead to non-volatile strengthening (LTP) and weakening (LTD) of synapses^[34,48,49].

To characterize this learning rule, spiking waveforms corresponding to SI-Note-3 Figure:S10 are applied to the pre- and post-synaptic terminals of the transistor as indicated, and the synaptic weight change is observed^[34,48,49]. For example, from figure 4A, an interval ($t_{\text{post-pre}}$) of +500ms result in the light pulse corresponding to a gate voltage of +6V, triggering a persistent increase in the channel conductance (LTP ~ 30%). Likewise, an interval of ($t_{\text{post-pre}}$) of -500ms results in the light pulse corresponded to a gate voltage of -6V, triggering a permanent decrease in the channel conductance (LTD ~ -25%) (SI-Note-3 Figures:S7-A). Synaptic weight change, the strength between two neurons is calculated using the relation, $\Delta W = (I_{\text{pre}} - I_{\text{post}}) / I_{\text{pre}}$, where I_{pre} and I_{post} are the currents of pre- and postsynaptic terminals and plotted as shown in figure 4A-D. Thus, with the existing single synaptic architecture, emulation of multiple forms of STDP improves the efficiency of storage and processing capacity in artificial neural networks. Different patterns of STDP learning rules have broader impacts on neural information processing in the field of neuroscience. Furthermore, the waveforms of the synaptic stimulus can be modulated for tuning the learning rules as demand.

Linear non-volatile blind updates: Unlike the asynchronous, univalued spikes of the SNNs, the neuron outputs in a deep neural network [DNN] are real-valued numbers that are processed in synchronous time steps^[50]. DNNs demonstrate tremendous potential for online machine learning weight updates with objective function minimizations implemented via backpropagation algorithms, labelled datasets, and highly parallel matrix multiplications. These networks need devices with a wide dynamic conductance range and conductance linearity (CL) characteristics, which can be modified through blind programming schemes^[51].

Here, we demonstrate electro-optical non-volatile conductance states, appropriate for analog computations. As shown in figure 4E, 20 input train of optical pulses with pulse width=0.05s, interval=1s, $\lambda=445\text{nm}$, and intensity = 12 mW/cm² are applied. Upon light illumination, the

conductance increases gradually (potentiation) which exhibits excellent non-volatility. Consequently, 20 electrical pulses are applied to drive back the trapped electrons and the conductance is reduced (depression). The potentiation-depression characteristics results in a near-linear manner, resulting in a reasonably symmetric response map, desirable for DNNs. To exhibit the stability of our devices, we have repeated this measurement for 10 cycles and observed nearly less conductance variation between the 1st and 10th cycle. i.e., 20 write pulses and 20 erase cycles for 10 consecutive cycles represents 400 switching transactions. This cycle-to-cycle reproducibility shows high stability, endurance and switching uniformity.

Figure 4. Electro-Optical programming of synaptic weights. Emulation of Hebb's STDP rules using optical presynaptic (blue light 445 nm) and electrical post-synaptic pulses. (A) anti-symmetric Hebbian, (B) anti-symmetric anti-Hebbian, (C) symmetric Hebbian and (D) symmetric anti-Hebbian learning rules for SNNs. (E) Linear Analog Non-Volatile Weight Updates accessed utilizing synergistic electrical and optical pulses (blue light – 445 nm) as shown in the inset of the zoomed-in figure. The graph on the left represents the endurance of our

devices across 400 switching cycles using blind updates and the graph on the right shows the zoomed-in view of the same (F).

Associative Learning: Classical conditioning experiments (Pavlov's dog) is finally conducted by coupling electro-optical pulses to demonstrate associative learning (Figure:5). Proposed by Ivan Pavlov, associative learning is an important tool for linking two events, i.e., a weak stimulus can be strengthened when it is concurrently associated with a strong stimulus^[52]. Through this pairing, the weak stimulus would respond like the strong stimulus and this technique could impact unique learning in the human brain. The optical pulses mimic unconditioned stimuli (US)/food triggering salivation/unconditioned responses (UR) while electrical voltage pulses trigger conditioned stimuli (CS)/bell for the artificial synapse (considered as "dog") activating conditioned responses (CR). Our devices depicts an UR (analogous to salivation) on noticing food/US (stage B, stimuli of 15 optical pulses, $\lambda = 445$ nm, pulse width=500ms, interval=500ms and intensity = 8 mW/cm²), marked by a high output resting current, above 10nA (threshold for salivation response). On the other hand, CS/bell ringing (stage A, stimuli of 15 voltage pulses, pulse width=500ms, interval=500ms and amplitude=2V) itself does not induce any salivation (current<10nA). Upon concurrent training with both US and CS or in other words simultaneous feeding and ringing the bell, the PSCs increases above the salivation response threshold (stage C) but failed to inculcate association. However, on repeated rehearsals (15 cycles), an effective association is developed in our devices, with the PSCs reaching well above the salivation threshold even upon removal of the US (stages D). This association remained valid for ~30 minutes but became extinct afterward, analogous to forgetting of the brain (stage E). Nonetheless, the association could be recovered by re-training as shown in stage F, establishing a robust association between the two modes of input stimuli.

Figure 5. Schematic of the Classical Conditioning experiments (Pavlov's dog experiment). Before conditioning, the artificial synapse (Pavlov's dog) produced significant outputs (salivated) only to US (stage: B) not to CS (stage: A). Note: A drain current (I_{ds}) of 10nA is set as the threshold for salivation. Persistent training with concurrent US and CS results in very high channel conductance, after which the artificial synapse (Pavlov's dog) produced significant outputs (salivated) in response to CS, indicative of associative learning (stages: C-D). The association weakened and became extinct with time (stage: E) but recovered upon retraining (stage: F).

Conclusion:

In conclusion, we have demonstrated a sensitized neuromorphic transistor that allows multi-level programming and linear conductance states via electro-optical gating. Type II heterojunction aids in realizing light-induced charge trapping (optical mode) and electric-induced charge releasing (electrical mode) properties aids in simultaneous optical programming and electrical erasing behaviour at the system level. Learning rules such as STDP, associative learning, and linear non-volatile blind updates are studied systematically to comprehensively demonstrate their compatibility with SNN and DNN computing architectures. The existing photosensitizer (CsPbBr_3) is sensitive to above bandgap illumination but remains insensitive to lower energies. The proposed IGZO synaptic device can acts as a fundamental device with a variety of photosensitizers such as dyes, organic semiconductors, 2D materials, and chalcogenide quantum dots. For example, utilizing perovskite quantum dots with lower bandgaps such as Cesium lead iodide (CsPbI_3) with bandgap $E_g \sim 1.7$ eV would allow for activation in the visible regime but not in infrared. In

comparison to pure electrical artificial synapses, such electro-optical synapses pave way for intelligent optical computing systems with high bandwidths and wavelength-division multiplexing for non-von Neumann computing schemes.

Experimental Section: Please refer to the supporting information for details on the experimental procedure, device fabrication, and characterization.

Supporting Information: Supporting Information is available from the Wiley Online Library or from the author.

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