

Short-Term Bienenstock-Cooper-Munro Learning in Optoelectrically-Driven Flexible Halide Perovskite Single Crystal Memristors

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Abstract: The transition to smart, wearable, and flexible optoelectronic devices that communicate with each other and perform neuromorphic computing at the edge, is a major goal in next-generation optoelectronics. These devices are expected to carry out their regular tasks while being supported by energy-efficient, in-memory computations. In this study, a lateral flexible device based on cesium lead tribromide perovskite single crystals integrated with single-walled carbon nanotube thin-film electrodes is presented. It is demonstrated that the device follows the Bienenstock-Cooper-Munro theory of synaptic modification under hybrid optoelectronic stimuli. This biorealistic response paves the way for the development of hybrid organic–inorganic artificial visual systems.

the high energy efficiency of biological organisms compared to classical computers.^[6–9] Within this framework, memristors are functional resistive elements capable of simultaneously mimicking the behavior of both neurons and synapses. As a result, they are considered to be fundamental building blocks for next-generation neuromorphic computing hardware.^[7,8,10–13]

The complex nonlinear synaptic functionality of memristive elements plays a crucial role in emulating biological neural networks. One notable example is the Bienenstock-Cooper-Munro (BCM) rule for synaptic modification, which is

naturally realized in the visual cortex.^[14–17] Within the framework of BCM theory, it is shown that the response of the nervous tissue to a sequence of electrical pulses nonmonotonically depends on the pulse repetition frequency (**Figure 1e**). Initially, the response decreases at low frequencies, corresponding to synaptic depression. It then reaches a minimum point before increasing again as the frequency rises. Importantly, there exists a specific threshold frequency at which no synaptic modification occurs. This threshold is dynamically adjusted based on the

1. Introduction

Modern computers based on the classical Von Neumann architecture are reaching energy consumption limits in supporting artificial intelligence (AI) algorithms and Internet of Things (IoT) applications.^[1,2] Neuromorphic computing has emerged as an alternative paradigm inspired by the structure and operation of the biological brain.^[3–5] The complexity of neurons and their connections, and the high level of parallelism are the reasons for

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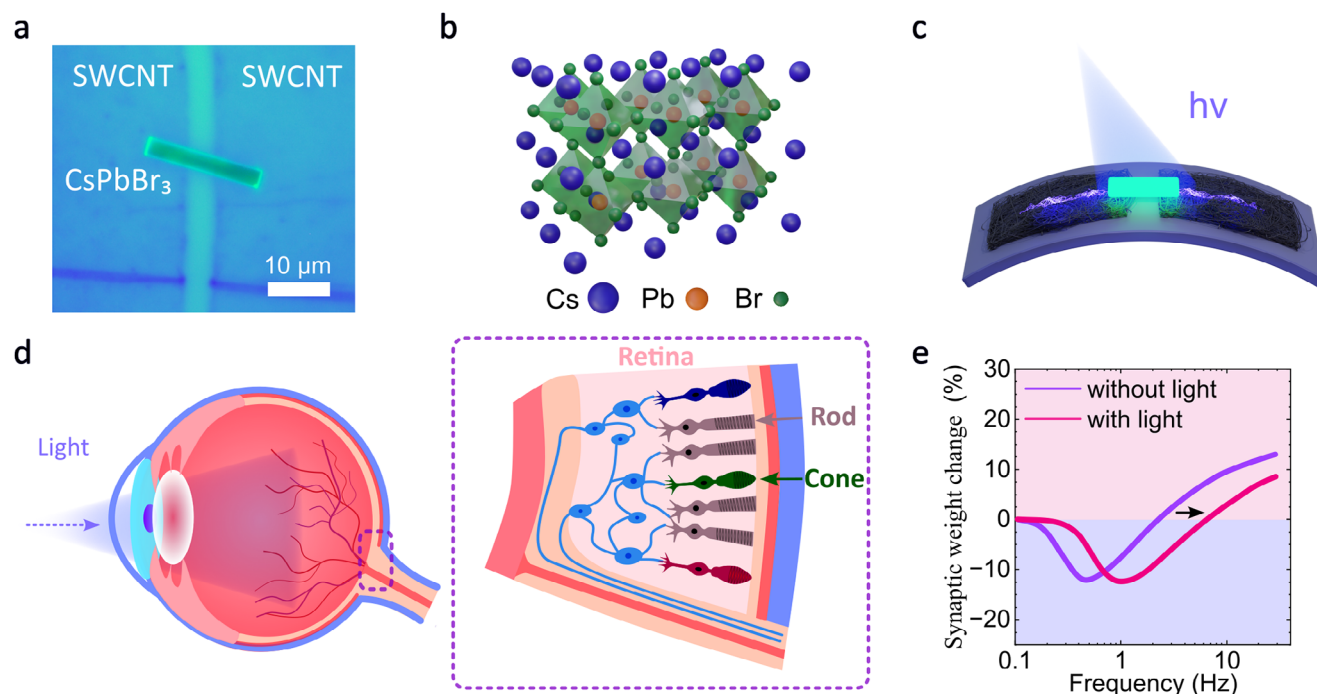


Figure 1. Visual system based on CsPbBr₃ single-crystal following the Bienenstock-Cooper-Munro rule. a) Microscopy image of a perovskite single-crystal on the flexible substrate connected to symmetrical SWCNT electrodes (photoluminescent image of the actual device). b) The orthorhombic crystal structure of the employed CsPbBr₃ microcrystal. c) Operation of the single-crystal device under combined optoelectrical stimuli and mechanical bending. d) Schematic of the human visual system: light is transmitted to the retina, where light-sensitive cones and rods send a collective response signal to the brain. e) Biological response of the device under hybrid optoelectrical stimuli, following the BCM rule. The synaptic weight change is frequency-dependent, exhibiting a depression region at low frequencies, followed by facilitation at high frequencies of internal input pulses. The sliding threshold, which separates depression and facilitation, is influenced by light stimuli.

characteristic intensity of light in the living organism's environment—specifically, how actively these brain regions are engaged in processing visual information.^[14,18,19]

Memristive devices implementing this learning mechanism require at least second-order dynamic behavior^[14,20] and can serve as functional components in neuromorphic hardware for image recognition tasks.^[20–22] Moreover, incorporating the BCM learning rule in spiking neural networks (SNNs) has been shown to significantly reduce training time,^[23] enhance network sparsity (thus improving energy efficiency),^[24] and outperform conventional spike-timing-dependent plasticity (STDP) models in SNNs with high neuronal connectivity.^[25] Ultimately, the BCM learning rule plays a pivotal role in advancing neuromorphic computing and enabling new approaches to real-time learning in artificial neural networks.

It is important that the BCM rule be implemented as directly as it is in living organisms to simplify the realization of neural network algorithms. Current research demonstrates BCM functionality in two different ways (Table 1). The first approach involves the preliminary stimulation of a memristor using rate-based spike trains that emulate synaptic activity under different sensory conditions. However, in this case, the synaptic weight change remains monotonic across the entire frequency range^[13,26,27] lacking the characteristic synaptic depression region.^[14] To address this limitation, a triplet spike-timing-dependent plasticity (triplet-STDP) scheme has been designed.^[20,21] Here, a sequence of three voltage pulses where one of the pulses differs in polarity from

the other two is received at the input of the device. Depending on the pulse repetition frequency (time intervals between spikes), the conductivity value of the device changes according to the BCM learning rule. Thus, a more direct BCM implementation that also incorporates a response to light stimuli, similar to that observed in the visual cortex of biological organisms, would be highly beneficial for further device development. This approach would allow computing tasks related to image recognition (involving a light source) to be processed naturally in-material,^[28,29] leveraging the advantages of the optical computing paradigm.^[30]

Table 1. Realization of BCM functionality in memristor devices.

Materials	Operation	BCM method	Timescale	Report
MAPbI ₃	Electrical	Triplet STDP	Long-term	[20]
WO _{3-x}	Electrical	Triplet STDP	Long-term	[21]
STO, Nb-STO	Electrical	Priming pulses	Short-term	[27]
CsFAPbI ₃ , CsPbBr ₂ I	Optoelectrical	Triplet STDP	Long-term	[31]
α-IGZO	Electrical	Priming pulses	Short-term	[32]
α-IGZO	Optoelectrical	Priming pulses	Short-term	[33]
ITO/GEL-CCs/ITO	Electrical	Priming pulses	Short-term	[34]
ITO/λ-car/ITO	Electrical	Priming pulses	Short-term	[35]
CsPbBr ₃ single crystal	Optoelectrical	Direct	Short-term	This work

Metal halide perovskites are a decent class of semiconductor materials exhibiting intrinsically complex charge carrier dynamics. Previously, they have been comprehensively studied aiming at not only optoelectronic applications such as photodetectors and solar cells,^[36–38] but also for memory applications,^[39–43] due to their dynamic nature of defects and mobile ions.^[20,39,40,44,45] Noticeably, the signature of the 2nd-order dynamics combined with the BCM learning rule has been revealed in multilayer device based on methylammonium lead iodide (MAPbI₃) perovskite by John et al., where the triplet spike-timing-dependent plasticity scheme was realized^[20] (Table 1). By integrating both memory functionality and light responsiveness, metal halide perovskites emerge as a promising material platform for the development of optoelectronic visual systems.

In this work, we demonstrate the short-term BCM response of cesium lead tribromide halide perovskite (CsPbBr₃) microcrystals to the hybrid optoelectrical stimuli. The photosensitive microcrystals are electrically connected to single-walled carbon nanotube (SWCNT) thin-film electrodes on a flexible substrate. These microcrystals mimic the cones embedded in the flexible human retina (Figure 1). Our results establish that in these devices, both frequency-dependent potentiation and depression, observed in both dark conditions and under light illumination, follow the Bienenstock-Cooper-Munro (BCM) learning rule. Moreover, we demonstrate that this BCM functionality, particularly in the depression region, does not require specialized electrical pulses in the form of triplets or presynaptic rate-based pulse trains (Table 1). Such a behavior upon hybrid optoelectronic stimuli can be explained by the competition of the capacitive- and inductive-like contributions coexisting in the charge carrier dynamics of metal halide perovskites.

2. Results and Discussion

Our study focuses on the lateral metal-semiconductor-metal type of the optoelectronic device.^[36,44] In such geometry, we can directly probe the dynamics of halide perovskite semiconductors, as compared to the typical vertical device structure with additional charge transport layers. To enable such a structure, we use a chemically inert SWCNT thin film as an electrode material, and CsPbBr₃ orthorhombic perovskite single-crystal as the semiconducting functional material (Figure 1a–c), similar to our previous works.^[36,44] When assembled on a flexible substrate, our crystals can be viewed as artificial cones embedded in the retinal layer of the human eye (Figure 1d). An additional advantage of halide perovskite single crystals over polycrystalline samples is their superior charge transport properties,^[36,46–49] as demonstrated by recent advancements in large-scale integration technologies for optoelectronic vision applications.^[50–53]

The fabrication of the CsPbBr₃ single crystal memristor consists of the following stages. First, we prepare nanostructured Al₂O₃ substrate with island-like morphology^[54] (Figure 2a,b; Figure S1a–e, Supporting Information). Then we conduct a solution-processed synthesis similar to the previous work.^[55] A small droplet (2 μ l) of metal halides (CsBr and PbBr₂) solution is dropped on the Al₂O₃ substrate and dried in the presence of 2-propanol-water azeotrope (H₂O-IPA) at 70 °C for 15–20 min (for details, see the Experimental Section). Thereafter, plenty of microcrystals are formed on the substrate (Figure 2c;

Figure S2a,b, Supporting Information). Scanning electron microscopy (SEM) image shows that single crystals are deposited on top of islands and have regular rectangular shape (Figure 2d; Figure S2b, Supporting Information). The photoluminescence spectrum of the microcrystals contains a single peak centered at 524 nm (Figure 2e). X-ray diffraction (XRD) pattern collected from an ensemble of microwire crystals in $\theta - \theta$ geometry is represented by two peaks corresponding to crystallographic planes (110) and (220), that matches with the reference data for the orthorhombic phase (space group P_{bmn})^[56] and confirms their directional growth (Figure 2f). Scanning transmission electron microscopy (STEM) image of a nanowire crystal grown along [001] axis (Figure 2g) reveals its monocrystalline structure. Energy-dispersive X-ray spectroscopy (EDX) images demonstrate the uniform distribution of Cs, Pb, and Br in the nanowire (inserted image in Figure 2g). The distance between parallel planes is 0.58 nm, which corresponds to crystallographic planes (002) of the orthorhombic phase of CsPbBr₃. Fast Fourier Transform (FFT) image obtained from Figure 2h contains sharp and bright points assigned to the planes (112), (110), and (002), indicating the periodic crystal lattice of CsPbBr₃ (Figure 2i). To confirm the monocrystallinity of the perovskite microwire, selected area electron diffraction (SAED) was performed (see Figure S3, Supporting Information). The SAED pattern in Figure S3b (Supporting Information) exhibits a series of sharp diffraction spots, including those corresponding to the (112), (110), and (002) planes.

To fabricate the device electrodes, the single-walled carbon nanotube (SWCNT) thin film is dry-transferred^[57] onto a flexible polyethylene naphthalate (PEN) substrate (see Figure S4, Supporting Information). The SWCNT film is then patterned into two separate electrodes (Figure 2j) by ablating a few-micrometer-wide path using a femtosecond laser^[36] (for details, see the Experimental Section; Figures S4,S5, and S6, Supporting Information). To complete the device fabrication, the as-synthesized microwire crystal (Figure 2k) is dry-transferred onto the prepared SWCNT film electrodes (for details see Experimental Section and Figures S7 and S8, Supporting Information).

As a result, we obtain a lateral SWCNT-CsPbBr₃-SWCNT structure of metal-semiconductor-metal (MSM) type (Figure 2l; Figure S9, Supporting Information), forming symmetrical Schottky contacts.^[44,58,59] The interelectrode distance of about 5 μ m (Figure 2l, inset image; Figures S6 and S9, Supporting Information) is comparable with the charge carriers diffusion length^[60–62] of the halide perovskite semiconductors, which is desirable for efficient charge carrier extraction and subsequent improvement of the device performance. To validate the stability of the formed contact between SWCNT and CsPbBr₃, we applied optoelectrical stimuli to the device and measured the response for each of 5000 bending cycles. As a result, after 20000 bending cycles, the device response decreased to 85% of its initial value (Figure S10, Supporting Information), thus manifesting the stable electrical contact of the as-formed structure.

Current–voltage measurements on the device in the dark and under continuous wave (CW) light illumination of 30 mWcm^{−2} at 532 nm wavelength are shown in Figure 2l (Figures S11 and S12, Supporting Information; for a detailed description of the optoelectrical setup see Experimental Section). The hysteresis I – V curves in the dark have an inverted inductive-like response, where the current on the reverse scan is higher than that of the

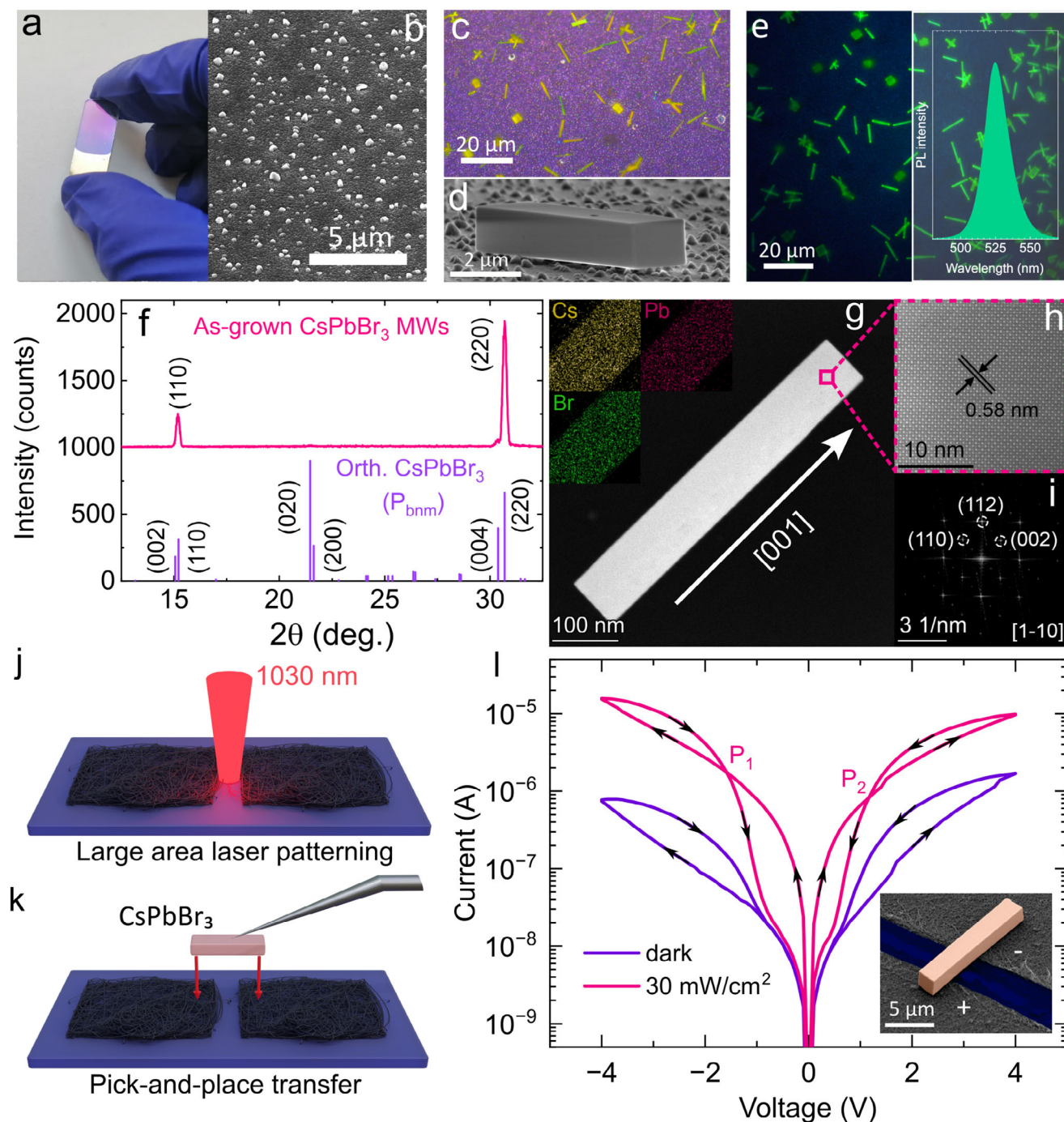


Figure 2. Fabrication of perovskite microcrystals, characterization of their optical and structural properties, and manufacturing of a memristor device. a) Macroscopic image of the nanostructured Al_2O_3 substrate. b) SEM image of island-like morphology of the Al_2O_3 substrate. c) Bright-field image of CsPbBr_3 microcrystals deposited on the substrate. d) Tilted-angle SEM image of a single-crystal on top of Al_2O_3 islands. e) Photoluminescent image and spectrum of the microcrystals. f) XRD pattern for an ensemble of as-grown CsPbBr_3 microwire crystals and the reference pattern. g) STEM image and EDX mapping of an isolated CsPbBr_3 nanowire crystal. h) High-Resolution STEM image of a selected area (indicated by a pink square on the (g)). i) FFT image obtained from (h). j) Illustration of femtosecond laser cutting off SWCNT thin film into two symmetrical electrodes. k) Dry transfer of a single crystal over SWCNT thin film electrodes. l) Current-Voltage curve in the dark (purple curve) and under illumination (pink curve). P_1 and P_2 are points where the hysteresis type changes from the regular to the inverted one. Arrows indicate the voltage sweeping direction. The inset picture represents the SEM image of the as-fabricated device.

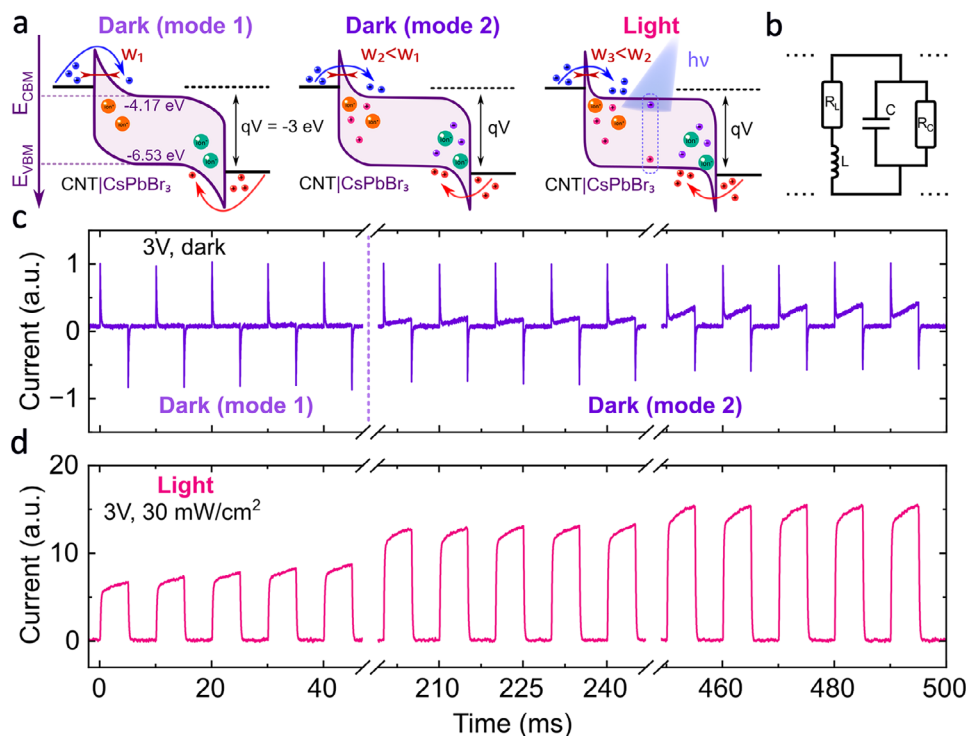


Figure 3. Transient dynamics and equivalent scheme approach. a) The schematic electronic band diagrams are the corresponding parts of the electrical circuit equivalent scheme b) and type of the transient dynamics. Mode 1 in the dark case corresponds to initial pure capacitive regime which transforms to mode 2, when ionic component starts growing. Under hybrid optoelectrical stimuli for 3 V pure inductive regime is implemented. On the diagrams big orange and green points represent intrinsic perovskite mobile ions, coexisting together with injected electrons (blue) and holes (red), and with photogenerated electrons and holes in case of light illumination. W_i denotes the width of Schottky barrier. The transient dynamics (3 V, 5 ms pulses) of perovskite memristor in the dark case (c) and under hybrid optoelectrical stimuli (d), respectively. The current is normalized to the maximum value of dark current.

forward scan.^[63] This behavior is characteristic of volatile memristors and is attributed to Schottky contacts modulated by mobile halide perovskite ions.^[42,44,64,65]

Under light illumination the hysteresis becomes more complex (Figure 2l). At low voltages, it exhibits a capacitive-like response (where the current on the forward scan is higher than that of the reverse scan), while at higher voltages, it transitions to an inductive-like response. The transition between these two regimes is indicated on the plot as P_1 and P_2 points (around -1.6 and 1.2 V, respectively). Such complex hysteresis transitions are commonly observed in halide perovskite optoelectronic devices with various structures.^[66–72] Such hysteresis transition in Figure 2l^[69,71] is attributed to the coexistence of inductive and capacitive types of response in perovskite devices.^[66,73] It occurs due to the combination of large surface capacitance of the perovskite, and the delay of recombination current, both due to ionic effects. Specifically, at low voltages, the polarization effect dominates, leading to capacitive behavior. However, at higher voltages, the electronic carriers cannot follow the ionic motion, giving rise to an inductive response. This behavior has been consistently reproduced in ten different single-crystal memristors (see Figure S18, Supporting Information). Therefore, our CsPbBr₃ MSM memristor contains at least two competing processes that can be qualitatively described as capacitive and inductive circuit branches (see Figure S13, Supporting Information).

Building on this observation, we further investigated how the voltage scan rate affects the hysteresis transition points. Notably, increasing the scan rate shifts the hysteresis transition points to higher (absolute) voltages. In Figure S14 (Supporting Information), we show that for a similar sample, increasing the scan rate from 0.5 to 10 V s⁻¹ shifts the hysteresis transition voltage from 2 V to over 3 V. Beyond 60 V s⁻¹, the hysteresis undergoes a transformation: the transition from capacitive-like to inductive-like behavior disappears, and the device exhibits a purely capacitive-like response. At even higher scan rates, the hysteresis gradually vanishes and eventually becomes purely inductive-like. These results show that the conductive and inductive behavior of the device can be easily controlled by adjusting the voltage scan rate, allowing for tunable hysteresis responses.

To explain the transient dynamics of the competing dynamic processes we provide the schematic electronic band diagrams of the device and the equivalent electrical circuit. The memristive behavior of our device is attributed to the modulated Schottky contacts by the intrinsic perovskite ions, such as bromide ions and their vacancies^[44]. The accumulation of the mobile ions at the interfaces leads to electrostatic lowering and shrinking of the energy barrier at the carbon nanotube-perovskite interface (Figure 3a). In the equivalent circuit this can be represented by the inductive branch (resistor and inductor in series). Because the inductive component is attributed to the slowly moving ions,

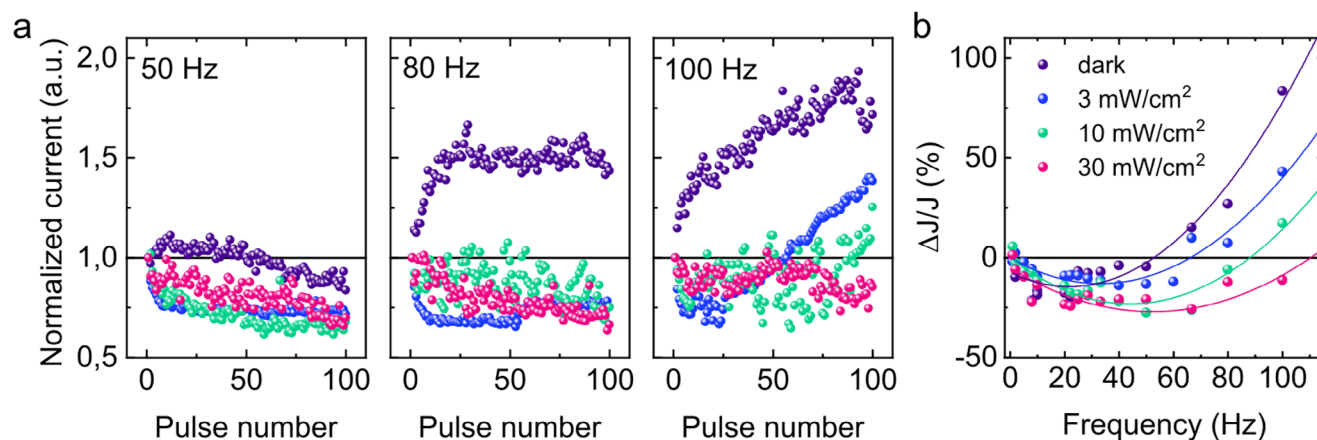


Figure 4. Experience-dependent synaptic plasticity featuring short-term Bienenstock-Cooper-Munro learning under hybrid optoelectrical stimuli. (a) Current response (normalized to the initial current) to 100 consecutive pulses (2.5 V, 5 ms) with different frequencies under electrical and optoelectrical stimuli. (b) The synaptic weight change inside a sequence of 100 pulses versus their frequency shows the “tick-like” shape similar to the BCM theory. The plot is separated into two parts for positive and negative relative change in synaptic weight. The light intensity shifts the zero point of the curve, showing the sliding threshold property. The solid lines indicate parabolic fit.

their appearance in the transient dynamics can be much slower, compared to the capacitive response of the electronic nature. Following the application of the 3 V bias one can see the slow change from the capacitive to inductive dynamics, manifesting the change of the Schottky barrier (Figure 3c).^[74] The parallel connection of the inductive and capacitive circuit branches highlights the competition between two contributions to the transient current response (Figure 3b) (see also Figure S13, Supporting Information). A similar scheme has been successfully validated in the literature describing halide perovskite optoelectronic devices.^[63,66,69–71,75–77]

Under light illumination many additional processes may be activated in halide perovskites. It includes the generation of photo-carriers, carrier trapping, heating, chemical reactions, etc.^[40,78,79] The huge modulation of the current across the device under light illumination (Figure 2l), means that the photoconductive effect dominates the transient response of the device under hybrid optoelectrical stimuli (Figure 3a).^[36] Here, the nature of the Schottky interface allows the photogenerated carriers to additionally contribute to the modulation of the Schottky barrier^[44] thus increasing the overall current and resulting in the inductive-like behavior of the device (Figure 3d).

2.1. Short-Term BCM Response

For a deeper investigation of the dynamical synaptic behavior of our memristors, we applied trains of 100 voltage pulses with an amplitude of 2.5 V and a pulse width of 5 ms. We monitored the current response as a function of pulse train frequency and light illumination intensity (Figure 4a). We observed that the current evolves non-monotonically over the course of 100 voltage pulses.

In the dark, at high frequencies (100 Hz), the current initially rises rapidly, following an inductive-like transient response, before transitioning into a slower increase. At intermediate frequencies (80 Hz), the current stabilizes, whereas at low frequencies (50 Hz), it may even decrease. Under varying light intensities (3 – 30 mWcm^{−2}), the current exhibits different behaviors:

at low frequencies, it decreases over the pulse train, indicating capacitive-like behavior, whereas at higher frequencies, it stabilizes or undergoes a steep increase. This inductive-like behavior can be attributed to the activation of ionic transport, which occurs alongside the relatively faster photogenerated electronic transport, also known as photoinduced capacitance.

It is important to note that the observed behavior, particularly following the +2.5 V pulse train at 100 Hz under illumination with the light power density of 30 mWcm^{−2}, which exhibits a capacitive-like transient response, is qualitatively different from that demonstrated in Figure 3d, where a similar +3 V pulse train results in inductive-like behavior. Despite this difference, in the *I*–*V* curve (Figure 2l), both +2.5 and +3 V fall within the same inductive-like branch. This counterintuitive discrepancy can be attributed to the significantly higher voltage scan rates in pulsed mode compared to quasi-DC *I*–*V* measurements in Figure 2l. As mentioned previously, higher voltage scan rates shift the hysteresis transition point toward higher voltages (Figure S14, Supporting Information), which we believe accounts for this effect. Such a sensitive transition between capacitive and inductive behavior can be leveraged for neuromorphic functional systems, as will be demonstrated below.

For a clearer comparison between different light intensities for each pulse train, we track the relative current change over the pulse train, calculated as:

$$\Delta J/J = (\langle J_{\text{last}} \rangle - \langle J_{\text{first}} \rangle) / \langle J_{\text{first}} \rangle \quad (1)$$

where $\langle J_{\text{first(last)}} \rangle$ represents the current averaged over the first (last) five pulses, plotted as a function of train frequency for various light intensities (Figure 4b). In our case $\Delta J/J$ serves as the measure of short-term synaptic plasticity of the optoelectronic memristor. This value governs the assignment of potentiation and depression regions to the areas above and below $\Delta J/J = 0$, respectively.

For our single-crystal memristors, we observe that for each light intensity the $\Delta J/J(f)$ dependence follows the same trend: starting from 0 (which corresponds to the idle situation,

obviously, not leading to any synaptic modification), the relative current change is negative and further decreases with frequency increase, reaching some minimum value and turning to the monotonic growth. Importantly, the value of the so-called threshold frequency (at which $\Delta J/J$ is equal or interpolated to 0) increases monotonically with the light intensity (Figure S15, Supporting Information). These findings closely align with the BCM theory of synaptic plasticity in the visual cortex.^[18,21,80] Since BCM plasticity plays a crucial role in visual pattern recognition, its direct integration into optoelectronic neuromorphic devices is highly valuable. Importantly, the reported device dynamics falls within a timescale range from milliseconds to seconds (Figure 3c,d; Figure 4a,b), thus aligning with biological timescales. Moreover, the studied light intensities correspond to natural visible light conditions, spanning from overcast or shaded environments to bright, direct sunlight. We note that our devices lose functionality after several hours of continuous operation, however, we do believe that proper encapsulation techniques will significantly extend device lifetime. Given the diversity of the perovskite materials and available fabrication methods, stable and efficient operation under realistic conditions looks achievable.

The effects of light illumination on the $\Delta J/J(f)$ dependence – namely, the increase of threshold frequency (Figure 4b) and the decrease in $\Delta J/J$ value at high frequencies – are analogous to the experience-dependent plasticity described by the BCM theory. In neurobiological systems, sensory deprivation during an organism's lifetime results in a sliding threshold frequency and increased electrical sensitivity in brain regions responsible for visual information processing. Notably, in our memristors, short-term BCM behavior is implemented directly, replicating classical neurobiological experiments without requiring complex preconditioning techniques such as preliminary electrical excitation at different frequencies^[13,26,32–35] or triplet STDP.^[20,21] This simplicity is advantageous for technological applications. The lack of long-term memory in our memristor does not reduce its potential for neuromorphic computing. In fact, its simple rate-based coding makes it well-suited for the use in a dedicated neuromorphic core that handles complex plasticity rules. This allows for faster synaptic weight updates, which can be stored in external non-volatile memristor cells, following the SpiNNaker architecture^[25] and IBM's PCM-based crossbar arrays.^[81]

3. Conclusion

In summary, we have investigated single-crystal CsPbBr₃ memristors assembled in a lateral geometry on a flexible substrate. These devices exhibit highly responsive photodetection (800 AW⁻¹ at 2.5 V of applied bias and under 30 mWcm⁻² illumination) while maintaining the performance under the mechanical deformation of 20000 cycles. Our devices exhibit biorealistic synaptic behavior, where light intensity dynamically adjusts the sliding frequency threshold in line with Bienenstock-Cooper-Munro theory. Importantly, the BCM functionality is achieved within a biologically relevant light intensity range of 1 mWcm⁻² – 30 mWcm⁻², aligning well with natural visual processing conditions. Furthermore, the modulation frequency of the BCM behavior spans the range from 0 to 100 Hz at the applied bias 2.5 V, mirroring the timescales of the biological systems. The observed

quantitative benchmarks establish that the perovskite material class represents a promising platform for realizing BCM learning in the presence of light. By integrating retinomorph (photodetecting) properties with visual cortex-inspired BCM synaptic behavior within a single halide perovskite microcrystal memristor, we introduce a material platform that could streamline visual information processing. Unlike both biological and conventional electronic systems, where sensing (photoreceptors/cameras), information transfer (neurons/interfaces), and processing (visual cortex/software) are distinctly separate, our approach suggests a more compact and efficient alternative. The in-sensor computing capabilities demonstrated in this work highlight the potential for seamless integration into neuromorphic chips for real-time visual data processing, paving the way for energy-efficient, hardware-based vision systems with reduced computational complexity.

4. Experimental Section

Fabrication of Substrates with Island-Like Morphology: To fabricate Al₂O₃ substrates with island-like morphology, a protocol was utilized similar to the previously reported one.^[54] First, a thin aluminum film was thermally deposited under vacuum on glass substrates. Then, the substrates were immersed in a 0.3 M aqueous solution of oxalic acid and anodized at 7 V for 15 min. Upon anodizing, the metallic film transformed into a non-conductive Al₂O₃ layer (Figure 2a,b; Figure S1, Supporting Information). Finally, the substrates were rinsed with deionized water to wash away acid moieties.

Synthesis of CsPbBr₃ Microcrystals: To obtain CsPbBr₃ microcrystals, a wet chemical synthesis was employed using a protocol similar to the one reported by Pushkarev et al.^[55] First, 0.1 M solution of stoichiometrically mixed metal halides (PbBr₂, 99.999%, Alfa Aesar, and CsBr, 99.99%, Sigma-Aldrich) was prepared in anhydrous dimethyl sulfoxide (DMSO, 99.8%, Alfa Aesar) in a N₂-filled glovebox. 2 μ L of the solution was drop-casted onto the Al₂O₃ substrate at ambient conditions and then the substrate was sealed in a hot Petri dish containing 250 μ L of H₂O-IPA azeotrope (95%) and dried on a hotplate at 70 °C for 15 min. As a result, numerous microcrystals distributed over the substrate with a mean density of 9.6×10^5 cm⁻² were grown and further utilized for a dry transfer procedure (Figure S2, Supporting Information).

Synthesis of SWCNT: Thin films of single-walled carbon nanotubes (SWCNTs) were produced using an aerosol (floating catalyst) chemical vapor deposition (CVD) method^[82] followed by a simple filtration via a nitrocellulose membrane (HAWP, Merck Millipore). For the aerosol CVD, ferrocene (98%, Sigma-Aldrich) was employed as a catalyst precursor while CO (99.99% Linde gas) acted as a carbon source. Ferrocene was transferred with a CO stream as a vapor to decompose upon reaching the hot zone (880 °C) of a quartz tube. The formation of Fe nanoparticles from ferrocene allows the catalytic Boudouard reaction ($2\text{CO} = \text{C} + \text{CO}_2$) to nucleate and produce nanotubes. Carbon dioxide (99.995% MGPZ) introduction tunes the nanotube properties (e.g., diameter distribution) and governs the process reactor productivity.^[83] The collected films consisted of individual SWCNTs exhibiting a mean diameter of ca. 1.9 nm, high quality, and random spatial orientation. After the deposition, SWCNTs form a randomly oriented uniform film suitable for further dry-transfer^[84] onto different types of a substrate.^[85]

Device Fabrication: SWCNT electrode preparation. The process of making electrodes from a SWCNT film consists of two stages. At the first stage, the film of 20 nm thickness on a cellulose filter paper was dry transferred to a polyethylene naphthalate (PEN) substrate cleaned with H₂O-IPA. After the removal of the filter paper, 20 μ L of H₂O-IPA was dripped to the transferred film to improve its adhesion to the substrate. Then, two droplets of silver paste (RS 186–3600) were deposited on the film and dried at the temperature $T = 70$ °C for 20 min. The droplets act as contact pads for

electrical connection (Figures S4 and S16, Supporting Information). Then, the sample was attached to double side tape on a glass substrate for further processing (Figure S4, Supporting Information).

At the second stage, laser ablation of the film to form interelectrode space was carried out using a fs laser (AVESTA ANTAUS-10W-4u/2.5M) with a pulse length of 270 fs, wavelength of 1030 nm, and pulse repetition frequency 4 kHz. Laser radiation with fluence of $\approx 2.3 \pm 0.5 \text{ Jcm}^{-2}$ was focused through a 50x objective (NIR Mitutoyo, NA = 0.42) on the sample surface at normal incidence (see Figures S5, and S6, Supporting Information).

Dry transfer of microwires. Perovskite microcrystals on the Al_2O_3 substrate could be dry transferred to the SWCNT electrodes by using an adhesive polydimethylsiloxane (PDMS) lens. The lens was prepared according to the following protocol. A silicon elastomer base (SYLGARD 184) and a curing agent (SYLGARD 184) were mixed in a 2:1 ratio by mass and stirred vigorously (see Figure S7a, Supporting Information). The resultant mixture was further degassed (see Figure S7b, Supporting Information), and a droplet of the mixture was deposited onto a glass substrate and kept at $T = 90^\circ\text{C}$ in an oven for 1 h. Note that the glass substrate with the droplet must be flipped upside down during the curing process to give a semi-convex shape of the lens that makes it possible to detach a single perovskite microcrystal from the Al_2O_3 substrate. The droplet was then completely solidified at room temperature for 12 h.

Thereafter, the prepared substrate with the PDMS lens was mounted on a three-axis positioner on an optical microscope (Zeiss Axio imager 2). The dry transfer procedure illustrated in Figure S8 (see Supporting Information) was conducted under 20x objective (EC Epiplan-Neofluar 20x/0.50).

Optoelectrical Measurements: To perform combined optoelectrical measurements the sample was mounted on Cascade Microtech Summit 11000M probe station. The probes were landed on Pt layers deposited onto two separated Si substrates, which in turn were connected with the SWCNT thin film electrodes via 50 μm gold wires and silver paste. Keysight B1500A semiconductor device analyzer was used for the electrical measurements. The optical stimuli were applied using CW 532 nm laser with the power density of 30 mWcm^{-2} . I - V curves were measured using the source-measure unit (SMU) of the Keysight B1500A semiconductor device analyzer in the quasi-DC mode with the staircase sweeps with minimum integration time per voltage point of 3 ms. Pulse measurements for BCM behavior demonstration were carried out using Keysight B1530A waveform generator/fast measurement unit (WGFMU) (see Figure S16, Supporting Information). All measurements were performed at room temperature. The BCM results were reproduced statistically by measuring at least three cycles for each data point in Figure 4b, the statistical results with error bars are provided in Figure S17, (Supporting Information).

Contact Stability Measurements: To perform a contact stability test a bending machine similar to the one described in the work^[86] was used. The substrate with a perovskite memristor was located on the lower cylinder, while the two upper ones bent and unbent it in the direction shown by the arrows in Figure S10 (Supporting Information). Curvature radius of the bent sample was $R = 2 \text{ mm}$.

After every 5000 bending cycles, the sample was removed from the setup and examined on optoelectrical circuit consisting of wave generator (Keysight 33600A), oscilloscope (Keysight DSOX6004A), and CW laser (Fianium Supercontinuum). The sample was subsequently electrically excited by 100 voltage pulses with the width of 5 μs and the amplitude of 3 V upon illumination by the CW laser with the wavelength of 532 nm and intensity of 30 mWcm^{-2} . The response (photocurrent) of the sample to this excitation measured through the shunt resistance $R = 1 \text{ MOhm}$ is presented in Figure S10a (Supporting Information). Subsequently, the dependence of the maximum current value on the number of bending cycles was recorded and is depicted in Figure S10b (Supporting Information).

Potential of Scalable Manufacturing of Planar Networks: To address the issue of scaling that is relevant for the engineering SSN consisting of number of microcrystals connected with each other in a series and par-

allel mode the following strategy is considered rational. An array of microcrystals can be formed by using the "liquid knife" approach.^[87] It utilizes micropillars with lyophobic sidewalls and lyophilic tops on Si substrate to split perovskite-precursor liquid film on insulating substrate into microdomains confining the nucleation and growth of single-crystal microplates of uniform size. Thereafter, laser-patterned SWCNT electrode arrays on a polymer film (e.g., PMMA or PDMS) can be physically laminated onto the obtained array of microcrystals to form MSM junctions.^[88] Photoexcitation of individual microcrystals within created network can be realized via spatial light modulator (SLM).

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

A.M., A.K., and I.M. originated the idea. A.G.N. and D.V.K. synthesized single-walled carbon nanotube thin films. R.P., I.M., S.A., and A.E. contributed to the fabrication of the memristors. I.M., A.K., A.M., N.S., performed optoelectrical measurements of memristors. A.M., I.M., A.Z., A.K., D.S., I.G.S., A.Y., analyzed the data. A.P., A.Z., J.B., and A.M. supervised the project. A.M., I.M., A.K., and A.P. wrote the original draft. A.M. managed the whole project. All authors reviewed and edited the manuscript. All authors contributed to the discussions and commented on the paper.

Data Availability Statement

Please confirm the selected template for your Data Availability Statement below, which will be published as part of your article if it is accepted for publication.

Keywords

carbon nanotubes, inorganic single crystals, memristor, metal halide perovskites, synaptic plasticity

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