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Light polarization as a control knob for resistive switching in chiral perovskite memory

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ABSTRACT

Resistive random-access memory (RRAM) based on metal-halide perovskites has attracted significant interest for neuromorphic and in-memory computing applications owing to its simple structure and tunable material properties. However, practical implementation is limited by stochastic filament formation and the lack of effective control parameters beyond electrical bias and light intensity. Here, we introduce light polarization as an independent control variable for resistive switching using a chiral perovskite RRAM device. Owing to the intrinsic circular dichroism of the chiral perovskite, right- and left-circularly polarized light (RCP and LCP) generate different photocarrier populations under identical illumination intensity, enabling deterministic modulation of switching behavior. As a result, the set voltage (V_{set}) is reduced from 0.15 V in the dark to 0.10 V and 0.05 V under RCP and LCP, respectively, accompanied by improved switching uniformity. Conduction mechanism analysis reveals that polarization-dependent photocarrier generation induces a transition from interface-limited to bulk-dominated transport, thereby stabilizing filament nucleation. In addition, the incorporation of bulky chiral ligands, which enhances chirality, leads to enlarged interlayer spacing that facilitates uniform Ag ion infiltration and suppresses stochastic filament formation. Finally, polarization-modulated synaptic characteristics are demonstrated, and artificial neural network simulations show enhanced learning performance. These results demonstrate that polarization-driven control of resistive switching can be effectively reinforced by structural engineering, establish light polarization as a new control knob for resistive switching, and provide a viable pathway toward multifunctional memory and neuromorphic applications.

1. Introduction

The rapid growth of data-intensive applications and artificial intelligence has exposed the fundamental limitations of conventional von Neumann computing architectures, particularly in terms of energy efficiency and data transfer bottlenecks [1–3]. To overcome these challenges, neuromorphic computing systems that mimic the parallel and energy-efficient operation of the human brain have attracted significant attention [4,5]. In this context, emerging memory devices capable of emulating synaptic functions are essential for implementing

hardware-based neural networks. Among them, resistive random-access memory (RRAM) has emerged as a promising candidate for next-generation non-volatile memory and neuromorphic computing systems due to its simple metal–insulator–metal structure, high scalability, and compatibility with crossbar array architectures [6,7]. Despite these advantages, practical implementation remains limited by the stochastic nature of filament formation and the lack of versatile control parameters for modulating switching behavior beyond electrical bias [8].

To address these challenges, material systems that provide additional

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degrees of freedom for modulating resistive switching are strongly desired, with optical approaches emerging as a particularly effective strategy [9,10]. Metal-halide perovskites have recently emerged as attractive candidates due to their strong light absorption, efficient photocarrier generation, and coupled ionic–electronic transport [11, 12]. These properties allow light irradiation to directly influence filament formation by reducing the effective energy barrier, leading to lower set voltage (V_{set}) and improved switching characteristics. However, in most reported systems, light primarily acts as an auxiliary energy source, and its effect is largely governed by intensity. This limits its role as an independent control parameter, as increasing intensity can induce material degradation, while insufficient intensity results in only weak modulation. Circularly polarized light (CPL) offers a fundamentally different approach by introducing a non-thermal optical degree of freedom. In chiral materials, CPL with opposite handedness induces asymmetric photocarrier generation due to circular dichroism (CD) [13]. Because filament nucleation in RRAM is highly sensitive to local carrier density and the associated energy barrier, polarization-dependent carrier generation provides a direct pathway for modulating switching characteristics.

Chiral metal-halide perovskites (CPVK) provide an ideal platform for this purpose due to their strong CD and favorable ionic transport properties. When combined with bulky organic ligands, these materials exhibit enhanced chiroptical response and enlarged interlayer spacing [14,15]. The latter facilitates ion migration and uniform distribution within the active layer, supporting stable resistive switching. However, while previous studies have explored chiral synaptic characteristics, the direct use of light polarization as an independent control parameter to modulate resistive switching behavior has not yet been demonstrated in perovskite-based RRAM systems [16].

Here, we demonstrate polarization-controlled resistive switching in a chiral perovskite RRAM device, where the handedness of circularly polarized light systematically modulates switching behavior. The

polarization-dependent photocarrier generation directly influences filament nucleation dynamics, enabling deterministic control of V_{set} without altering the illumination intensity. In addition, the enlarged interlayer spacing induced by bulky ligands facilitates uniform Ag ion infiltration throughout the perovskite layer, further improving switching stability. Finally, polarization-controlled conductance modulation is extended to synaptic functionality, demonstrating enhanced neuromorphic performance.

2. Results and discussion

2.1. Electrical characteristics of the chiral perovskite RRAM

2D perovskites, especially Ruddlesden-Popper (RP) phases, are attractive active materials for RRAM because their layered structure suppresses out-of-plane leakage while promoting electronic confinement, charge trapping, and ion migration [17]. These features facilitate resistive switching behavior, and their compositional and chiral tunability further provide an additional degree of freedom for device functionality. Leveraging these properties, a chiral RP perovskite RRAM device was fabricated using (1-(2-naphthyl)ethylamine) $_2$ PbI $_4$, where 1-(2-naphthyl)ethylamine (NEA) serves as a bulky organic chiral spacer, inducing strong chirality. To establish a reliable device platform for polarization-controlled operation, the electrical characteristics of the chiral perovskite RRAM device were first investigated. We used (R-NEA) $_2$ PbI $_4$ (R-CPVK) for the device fabrication. Fig. 1a presents a schematic illustration of the R-CPVK RRAM device, composed of an Ag top electrode (50 nm) and an Au bottom electrode (30 nm) with a layered R-CPVK active layer (600 nm) in between. This vertical metal-CPVK-metal structure enables efficient formation and rupture of conductive filaments under an applied electric field, which is essential for resistive switching behavior. Fig. 1b illustrates the crossbar array architecture, highlighting the potential for high-density memory

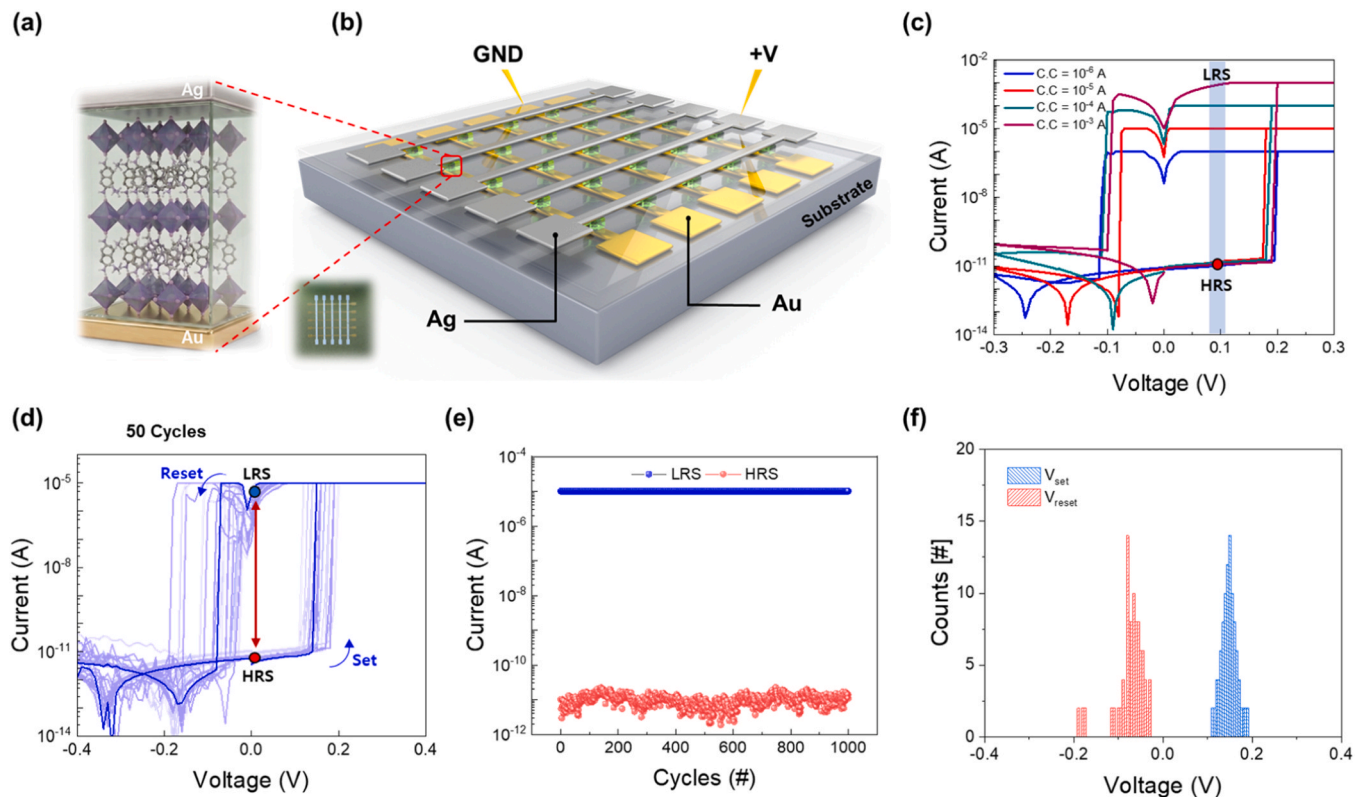


Fig. 1. (a) Schematic illustration of the Ag/(R-NEA) $_2$ PbI $_4$ /Au device structure. (b) Schematic illustration of the array-structured chiral RRAM. (c) I-V curves under difference compliance current. (d) I-V switching characteristics. (e) Endurance cycle test for 1000 cycles at a read voltage of 0.5 V. (f) Distribution of V_{set} and V_{reset} .

integration. Such a structure is particularly advantageous for neuromorphic computing applications, as it allows scalable implementation of synaptic devices with minimal footprint. To exploit the advantages of metallic filament-based switching, we employed an Ag top electrode to induce conductive-bridge RAM (CBRAM)-type operation. CBRAM devices are attractive because the electrochemical formation of metallic conductive filaments can provide low operating voltage, high resistance contrast, and large On/Off ratio. Consistent with this expectation, the devices show a markedly low set voltage ($V_{\text{set}} = 0.15$ V) and large On/Off ratio. In Fig. 1c, as the compliance current (CC) increases, the on-state current increases accordingly, while the off-state current remains nearly unchanged. As a result, the on/off ratio reaches approximately 10^8 under optimized CC conditions. This large On/Off ratio can be attributed to the suppressed off-state current, which originates from the incorporation of bulky organic ligands. The enlarged interlayer-spacing (d -spacing) enhances electronic confinement and inhibits charge transport in the out-of-plane direction. In addition, the intrinsically insulating nature of the NEA further increases the overall resistance of the active layers. Notably, the sharp transitions between the high-resistance state (HRS) and low-resistance state (LRS) suggest that the switching behavior is governed by the formation of conductive filaments, likely associated with Ag ion migration, consistent with a CBRAM mechanism [18,19]. To further examine the switching mechanism, a control device with an Au top electrode, which has significantly lower ion mobility than Ag, was fabricated. As shown in Fig. S1, the Au-based device exhibits switching behavior characteristic of vacancy-mediated conduction, commonly observed in perovskite-based RRAM systems [20]. Compared to the Ag-based device, the Au-based device shows higher operating voltages and a higher resistance in the low-resistance state (LRS), with an On/Off ratio of $\sim 10^3$ and V_{set} exceeding 1 V. These differences indicate that metal filament-based switching can provide more efficient switching characteristics than vacancy-driven conduction in terms of operating voltage and on/off ratio [21]. Fig. 1d shows that the I–V curves measured over 50 consecutive cycles exhibit nearly identical switching behavior, demonstrating excellent cycle-to-cycle repeatability. The device shows both the set and reset processes occur at well-defined voltages without noticeable fluctuation, indicating stable filament formation and rupture mechanisms. Fig. 1e shows that the device maintains stable switching operation over 1000 cycles with negligible degradation in resistance states. The resistance values of both the high-resistance state (HRS) and low-resistance state (LRS) remain clearly separated, confirming reliable switching stability. The statistical distributions of V_{set} and V_{reset} are presented in Fig. 1f. The extracted V_{set} value of $0.15 \text{ V} \pm 0.02 \text{ V}$ indicates a relatively narrow switching-voltage distributions under dark conditions. This result is notable because CBRAM-type filamentary switching is generally susceptible to stochastic variation, arising from the random nucleation, growth, and rupture of metallic conductive filaments [22, 23]. Despite this intrinsic limitation, the present device exhibits stable cycle-to-cycle switching and narrow voltage distributions, suggesting that the chiral RP perovskite provides a favorable environment for reproducible Ag filament formation. The possible origin of this improved switching uniformity, particularly the role of the enlarged interlayer spacing and Ag ion migration pathway, will be discussed in detail in the following section.

The optical and electronic structures of the chiral perovskite film were examined by UV–visible absorption and UPS measurements (Fig. S2). The optical bandgap was estimated to be 2.43 eV from the Tauc plot, and the VBM was determined to be 1.59 eV from the UPS valence band onset. Based on these values, the energy band diagram of the chiral perovskite was constructed (Fig. S3). The experimentally obtained band structure was consistent with the theoretical calculation, confirming the reliability of the extracted energy levels (Fig. S4). Notably, the optical bandgap is lower than the photon energy of the 488 nm laser used for CPL irradiation, suggesting that photocarrier generation can occur during polarization-dependent switching

measurements.

2.2. Polarization-controlled switching behavior of chiral RRAM

To move beyond conventional light-intensity-driven modulation, we investigate light polarization as an additional degree of freedom for controlling resistive switching behavior. To enable polarization-controlled operation, it is first necessary to verify the polarization-dependent optical response of the chiral perovskites. The CD spectrum of the (R-/S-NEA)₂PbI₄ is shown in Fig. 2a, confirming strong polarization-dependent optical absorption, with a CD intensity reaching ~ 300 mdeg. The corresponding UV-Vis absorption spectrum exhibits pronounced excitonic features in the same spectral region (Fig. S5), indicating efficient light absorption. Furthermore, the dissymmetry factor (g -factor) reaches ~ 0.011 , representing a strong chiroptical response. Such enhanced chiroptical activity is consistent with previous reports that bulky organic ligands can effectively amplify chirality in 2D CPVKs [15]. The distinct CD response indicates that RCP and LCP generate different carrier populations within the material under identical optical intensity. The influence of CPL irradiation on the switching behavior is presented in Fig. 2b. Under dark conditions, the switching occurs at 0.15 V, whereas under RCP and LCP irradiation, the switching voltage decreases to 0.10 V and 0.05 V, respectively. The reduction in V_{set} is attributed to photo-generated carriers, which lower the effective switching barrier for filament formation. Owing to the CD of the (R-NEA)₂PbI₄, LCP is absorbed more strongly than RCP, resulting in a high photogenerated carrier density. In addition, the current level in the HRS also increases, enabling multiple distinct current states. Notably, this multi-level behavior is also dependent on light polarization, further highlighting the potential of chiral perovskites for polarization-controlled memory functionalities. These results represent the first demonstration of polarization-controlled modulation of switching characteristics in a chiral perovskite RRAM device. The corresponding raw data for 25 switching cycles under dark, RCP, and LCP conditions are provided in Fig. S6, demonstrating the high reproducibility and reliability of the polarization-dependent switching behavior. To verify whether the CPL-dependent switching of the (R-NEA)₂PbI₄ based chiral RRAM is indeed dictated by its chirality, we fabricated control devices using (S-NEA)₂PbI₄ and evaluated their performance. In contrast to the (R-NEA)₂PbI₄ based chiral RRAM, the (S-NEA)₂PbI₄ based devices exhibited a pronounced decrease in V_{set} and a substantial increase in current specifically under RCP illumination (Fig. S7). These opposing characteristics clearly demonstrate that the polarization-dependent RRAM operation is driven by the molecular chirality of the perovskite layers. We calculated the photocurrent dissymmetry factors (g_{ph}) and photoresponsivities (R) of the R- and S-NEA-based RRAM devices under RCP and LCP illumination [24]. The results showed that the CPL-dependent photoresponse originates from the intrinsic chirality of the active materials. (Table S1 and S2 in Supporting information) While the two devices exhibit comparable g_{ph} values, the opposite signs of g_{ph} and the reversed photoresponse under RCP and LCP illumination demonstrate their enantioselective interaction with CPL. This conclusion is further supported by the polar plots of the photocurrent as a function of the azimuthal angle, which clearly reveal the chirality-dependent photoresponse of the devices. (Fig. S8) We used CPL at 488 nm with an intensity of 1 mW, consistent with our previous work, and attempted to investigate intensity-dependent conduction [24]. However, at lower intensities, no measurable switching behavior was observed, whereas at higher intensities, device degradation occurred. These results indicate that light intensity alone has inherent limitations as a control parameter. In contrast, CPL provides a unique degree of freedom for stable modulation of switching behavior. Recent studies have suggested that the CPL response in chiral perovskites is governed not only by CD, but also by the chiral-induced spin selectivity (CISS) effect. Owing to the broken inversion symmetry and strong spin–orbit coupling, Rashba-type spin splitting enables the

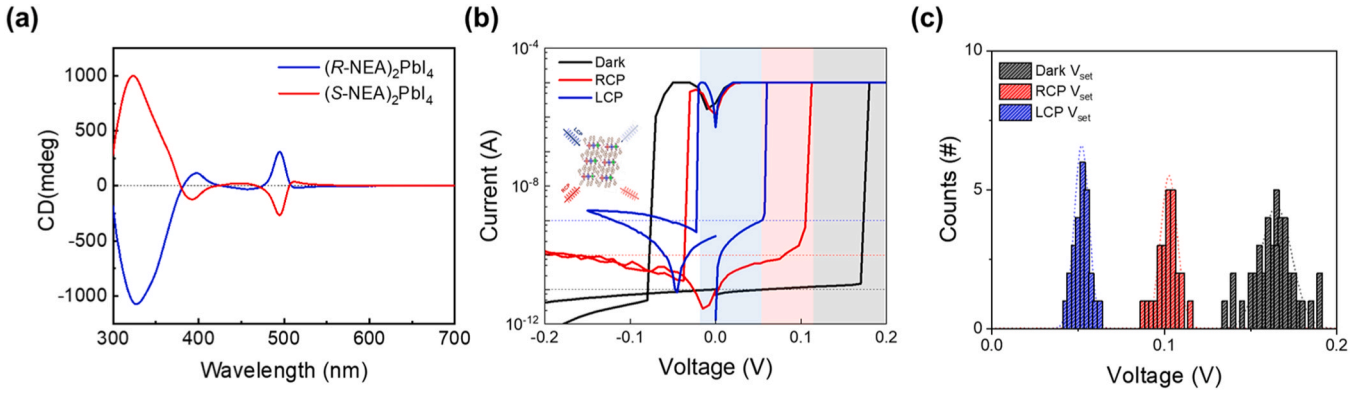


Fig. 2. (a) Circular dichroism of (R-/S-NEA)₂PbI₄. (b) I–V switching characteristics, (c) statistical distribution of V_{set} .

generation of spin-polarized carriers under CPL, while the CISS effect promotes spin-selective carrier transport [25,26]. Therefore, the observed CPL-dependent switching behavior is likely governed by the combined effects of spin-polarized carrier generation, spin-selective carrier transport, and spin preservation, rather than solely by polarization-dependent optical absorption. To support this interpretation, we further characterized the spin transport properties of the R- and S-NEA films (Fig. S9 and Fig. S10). The two enantiomers exhibit high spin polarization with opposite spin-filtering helicities, providing direct evidence that the CISS effect contributes to the observed CPL-dependent device response. The statistical distribution of V_{set} under different conditions is summarized in Fig. 2c. To evaluate the switching uniformity, the relative variation (standard deviation normalized by the mean, expressed as a percentage) was compared under different conditions. The extracted values are $\sim 9.2\%$, $\sim 5.9\%$, and $\sim 8.9\%$ for dark, RCP, and LCP, respectively. While the relative variation appears slightly smaller under RCP, this is mainly due to the reduced operating voltage, which can lead to an apparent underestimation of variability. Overall, the variation levels are comparable to those reported in previous studies [27,28]. Considering the absolute voltage fluctuations, both RCP and LCP conditions exhibit reduced variation compared to the dark state, indicating improved switching uniformity under CPL illumination. The preservation of the CPL-dependent response in the Au/R-NEA/Au control device indicates that the polarization selectivity is an intrinsic property of the chiral perovskite layer (Fig. S11). Combined with the Ag-infiltration control experiments, these results clearly separate the respective roles of the two mechanisms: Ag infiltration establishes a reliable switching platform, whereas the chiral perovskite provides the CPL-dependent modulation. These results demonstrate that light polarization serves as an effective and non-destructive degree of freedom for controlling switching behavior. Unlike energy-driven optical modulation, polarization-based control minimizes degradation while enabling reliable multi-level operation, along with precise tuning of V_{set} and switching uniformity. This approach provides a simple yet powerful alternative to complex multi-functional designs for achieving advanced switching functionalities.

2.3. Resistive switching mechanism analysis of chiral RRAM

To further understand the switching mechanism, conduction behavior was analysed. In our previous analysis, Schottky emission (SE) conduction was examined as a possible transport mechanism [19]. In this mechanism, electrons overcome the Schottky barrier formed at the metal–CPVK interface through a combination of thermal excitation and electric-field-induced barrier lowering. The SE conduction is expressed by Eq. (1).

$$J_{\text{SE}} = \frac{4\pi q m^* (kT)^2}{h^3} \exp \left[-\frac{q(\Phi_B - \sqrt{qE/4\pi\epsilon})}{kT} \right] \quad (1)$$

where, J_{SE} is the current density, q is the electron charge, m^* is the effective electron mass, k is Boltzmann's constant, T is the absolute temperature, h is Planck's constant, Φ_B is the Schottky barrier height, E is the electric field across the insulating layer, and ϵ is its dielectric constant. To verify this behavior, $\ln(I)$ versus $V^{1/2}$ plots were analysed under dark, RCP, and LCP conditions. As shown in Fig. 3a, the dark condition exhibits clear linearity, whereas the linearity becomes weaker under RCP and LCP irradiation, suggesting a deviation from pure Schottky emission behavior under illumination [29]. Further insight can be obtained from the $\ln(I)$ versus $\ln(V)$ plots shown in Fig. 3b. In the log-log analysis, the current can be expressed as $I \propto V^n$, where the exponent n is obtained from the slope of $\ln(I)$ versus $\ln(V)$ plot according to $\ln(I) = n\ln(V) + C$. Since ohmic conduction follows as $I \propto V$, a slope close to unity indicates a more ohmic-like transport behavior [30]. The extracted slope increases from 0.2 in the dark state to 0.58 under RCP and 1.02 under LCP irradiation, indicating a gradual transition from injection-limited transport toward more bulk-dominated conduction, inducing ohmic-like transport. Such analysis is commonly used to identify the dominant conduction mechanism, where an increase in slope reflects a transition from injection-limited transport to bulk-dominated conduction [31,32]. Under illumination, photo-generated carriers in perovskites are known to induce trap filling in both bulk and interfacial states, reduce interfacial barriers through screening effects, and facilitate carrier transport in the bulk [33,34]. Taken together, these results indicate that while the transport in the dark state is dominated by interface-limited injection, CPL irradiation introduces photogenerated carriers that reduce interfacial limitations and enhance bulk conduction through trap filling. This transition toward bulk-dominated transport is consistent with the observed stabilization of filament formation and may contribute to improved switching uniformity under CPL illumination. Notably, the characteristic trap-controlled ($n \approx 1-2$) and trap-filled ($n \geq 2$) SCLC regions are not observed prior to resistive switching. We attribute this behavior to the pre-existing uniform distribution of Ag within the perovskite layer, which allows conductive filaments to form once a modest increase in carrier transport is achieved, before conventional bulk-limited SCLC transport becomes dominant. Therefore, the CPL illumination primarily facilitates carrier transport and filament formation rather than inducing a classical SCLC conduction regime. For the LRS, as shown in Fig. 3c, the conduction behavior is nearly linear under all conditions. This is consistent with metallic-like conduction through a formed Ag filament, indicating that polarization primarily influences the filament formation stage rather than the fully formed conductive path. These results demonstrate that the polarization of light enables selective modulation of carrier transport pathways by distinguishing the handedness of incident light. This

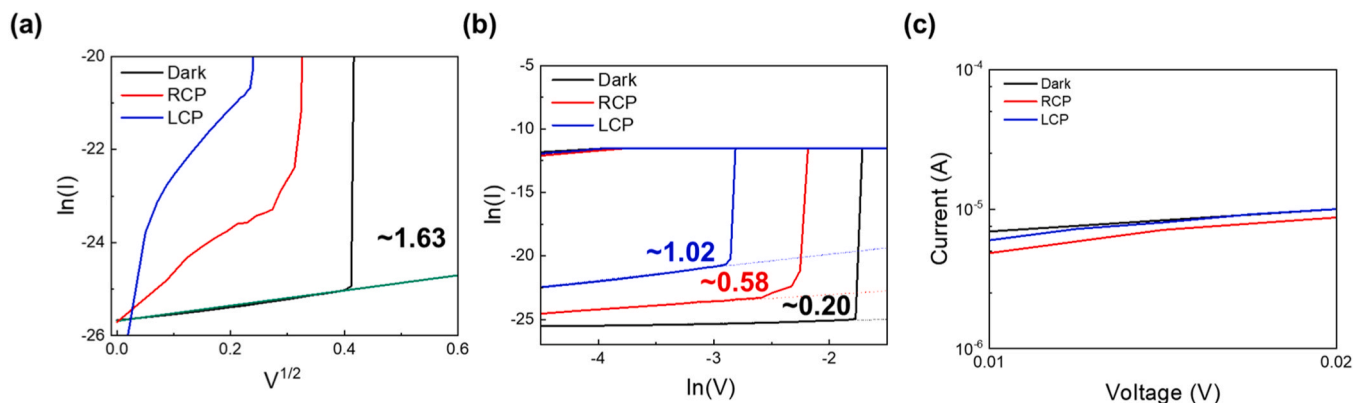


Fig. 3. (a) Schottky emission fitting of $\ln(I)$ vs $V^{1/2}$ (b) log-log I-V fitting of $\ln(I)$ vs $\ln(V)$, (c) log-scale fitting of LRS behavior of the chiral RRAM under dark, RCP, and LCP conditions.

polarization-dependent carrier generation effectively tunes the relative contribution of interface-limited and bulk-dominated conduction, providing a physical origin for the observed polarization-dependent switching behavior.

2.4. Structural origin of switching stability

To further elucidate the origin of the significantly improved switching uniformity observed under CPL illumination, it is necessary to consider additional factors beyond carrier modulation. While the conduction mechanism analysis indicates that polarization-dependent photocarrier generation can tune the balance between interface-limited and bulk-dominated transport, this alone does not fully account for the highly stabilized filament formation behavior. In previous studies, structural engineering strategies—such as the introduction of metal nanodots, nanoisland arrays, or localized dopants—have been employed to spatially confine filament formation and improve switching uniformity [35–37]. Motivated by these approaches, we investigated whether the structural characteristics of the chiral perovskite itself could provide an intrinsic origin for the enhanced uniformity. In particular, the bulky chiral ligand NEA, originally introduced to induce strong chiroptical activity, also results in an enlarged interlayer spacing in the $(R\text{-NEA})_2\text{PbI}_4$ structure. This expanded spacing is expected to provide more accessible diffusion pathways for Ag species, facilitating their penetration into the perovskite layer.

To experimentally verify Ag distribution, cross-sectional SEM imaging combined with energy-dispersive X-ray spectroscopy (SEM-EDX) was performed (Fig. 4a). The elemental mapping clearly shows that Ag is distributed throughout the $(R\text{-NEA})_2\text{PbI}_4$ layer, rather than being localized near the top electrode, indicating effective vertical infiltration. To further examine the spatial distribution, 3D TOF-SIMS analysis was conducted (Fig. 4b). The depth profile exhibits a relatively uniform Ag signal intensity across the perovskite layer, and the reconstructed 3D images clearly demonstrate that Ag is uniformly dispersed throughout the entire $(R\text{-NEA})_2\text{PbI}_4$ layer. Such uniform Ag infiltration, enabled by the enlarged d -spacing, is expected to provide well-distributed ionic migration pathways, thereby lowering the energy barrier for filament formation and leading to reduced V_{set} and improved switching uniformity. In resistive switching devices, ion transport and subsequent redox reactions play critical roles, where ion migration typically requires a high activation energy [38,39]. According to the field-assisted ion migration model, the migration velocity can be simply expressed as

$$v = v_0 \exp\left(-\frac{E_a - qEa}{kT}\right) \quad (2)$$

where v is the ion migration velocity, v_0 is the attempt frequency, E_a is the activation energy for ion migration, q is the ionic charge, E is the

applied electric field, a is the hopping distance, k is the Boltzmann constant, and T is the temperature [40,41]. This relationship indicates that the migration rate is exponentially governed by the effective activation energy, and thus reducing the migration barrier is essential for lowering the switching voltage. Building on this model, the pre-existing uniform infiltration of Ag within the $(R\text{-NEA})_2\text{PbI}_4$ layer effectively reduces the ion migration barrier, enabling filament formation at lower voltages, which is consistent with the observed reduction in V_{set} . In addition, the homogeneous spatial distribution of Ag ensures that filament nucleation occurs under similar local conditions throughout the device, leading to reduced stochasticity in filament formation and consequently enhanced switching uniformity. In contrast, $(R\text{-}(+)\text{-}\alpha\text{-methylbenzylamine})_2\text{PbI}_4$ exhibits limited Ag penetration due to its relatively narrow interlayer spacing, as confirmed by both SEM-EDX and TOF-SIMS analyses (Fig. S12 and S13), which show that Ag is primarily localized near the top electrode with minimal diffusion into the bulk region. Such restricted ion distribution leads to non-uniform and localized filament formation, resulting in higher operating voltages and poor switching uniformity [16]. In this case, an electroforming process is likely required to initiate conductive pathways, further increasing the variability of switching behavior. This difference in filament formation behavior between $(R\text{-NEA})_2\text{PbI}_4$ and $(R\text{-MBA})_2\text{PbI}_4$ is schematically illustrated in Fig. 4c, where the enlarged interlayer spacing in the NEA-based structure enables uniform Ag infiltration and well-distributed filament formation, whereas the limited spacing in the R-MBA-based structure restricts ion migration, resulting in localized and less stable conductive paths. These results demonstrate that ligand-engineered d -spacing expansion intrinsically enables homogeneous ion migration pathways without additional processing, establishing interlayer spacing as a key design parameter for achieving effective Ag infiltration and highly uniform resistive switching.

2.5. AIMD simulations of Ag diffusion in $(R\text{-NEA})_2\text{PbI}_4$ and $(R\text{-MBA})_2\text{PbI}_4$

To investigate the effect of ligand-engineered d -spacing on Ag migration, ab initio molecular dynamics (AIMD) simulations were performed for $(R\text{-NEA})_2\text{PbI}_4$ and $(R\text{-MBA})_2\text{PbI}_4$ structures with an Ag ion (Figs. 5a and 5b). As shown in Fig. 5c, the mean square displacement (MSD) of an Ag atom in $(R\text{-NEA})_2\text{PbI}_4$ is significantly larger than that in $(R\text{-MBA})_2\text{PbI}_4$ over the entire simulation time, indicating enhanced ionic mobility in the $(R\text{-NEA})_2\text{PbI}_4$. This increased mobility might be attributed to the larger organic ligand in $(R\text{-NEA})_2\text{PbI}_4$, which leads to an expanded interlayer spacing and a more flexible lattice framework. The AIMD trajectories (Movie S1 and Movie S2) further support that Ag-ion migration becomes easier in $(R\text{-NEA})_2\text{PbI}_4$ due to its enlarged interlayer spacing. The increased structural flexibility reduced the

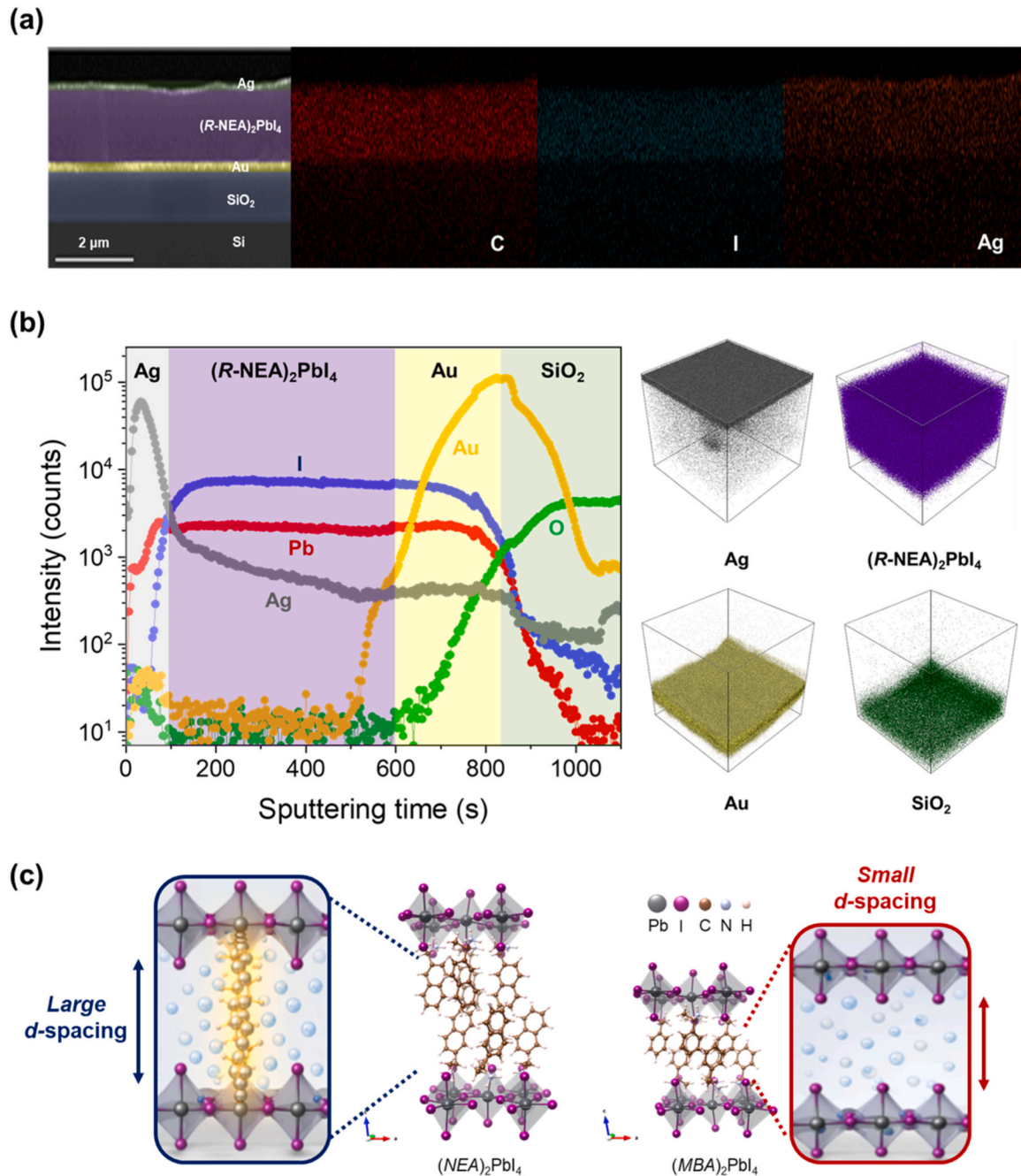


Fig. 4. (a) Cross-sectional SEM image and corresponding EDX elemental mapping and (b) 3D TOF-SIMS depth profiling of elements of (R-NEA)₂PbI₄. (c) Schematic illustration of crystal structures and *d*-spacing-dependent Ag distribution and filament formation in (R-NEA)₂PbI₄ and (R-MBA)₂PbI₄.

migration barrier for Ag ions, thereby facilitating their diffusion within the perovskite lattice. In contrast, the relatively smaller ligand in (R-MBA)₂PbI₄ results in a more rigid and confined structure, suppressing Ag migration. These computational results are consistent with the TOF-SIMS results (Fig. 4b), where Ag signals were observed in the perovskite region only for (R-NEA)₂PbI₄, confirming the experimentally enhanced diffusion of Ag in this structure. Therefore, the combined theoretical and experimental results suggest that ligand-induced structural modulation plays a critical role in governing Ag migration behavior. The facile migration of Ag in (R-NEA)₂PbI₄ promotes the formation of conductive Ag channels, which can act as additional charge transport pathways independent of photo-generated carriers, potentially contributing to device performance. The systems were equilibrated during the AIMD simulations, as confirmed by the stable fluctuations of temperature and

total energy (Fig. S14). Control devices containing a thicker PMMA interlayer exhibit a significantly higher V_{set} and poorer switching uniformity than the original devices with uniformly infiltrated Ag (Fig. S15). These results demonstrate that the enlarged interlayer spacing facilitates uniform Ag infiltration, which is essential for achieving low-voltage, highly uniform resistive switching. Therefore, engineering the interlayer spacing provides an effective materials-design strategy for realizing reliable and energy-efficient RRAM devices. However, the uniform distribution of Ag may also contribute to the short retention behavior. As shown in Fig. S16, the device with uniformly distributed Ag exhibits relatively short retention, whereas the device incorporating a PMMA interlayer to regulate Ag infiltration demonstrates significantly prolonged retention. These results indicate that controlling the degree of Ag infiltration provides an effective means

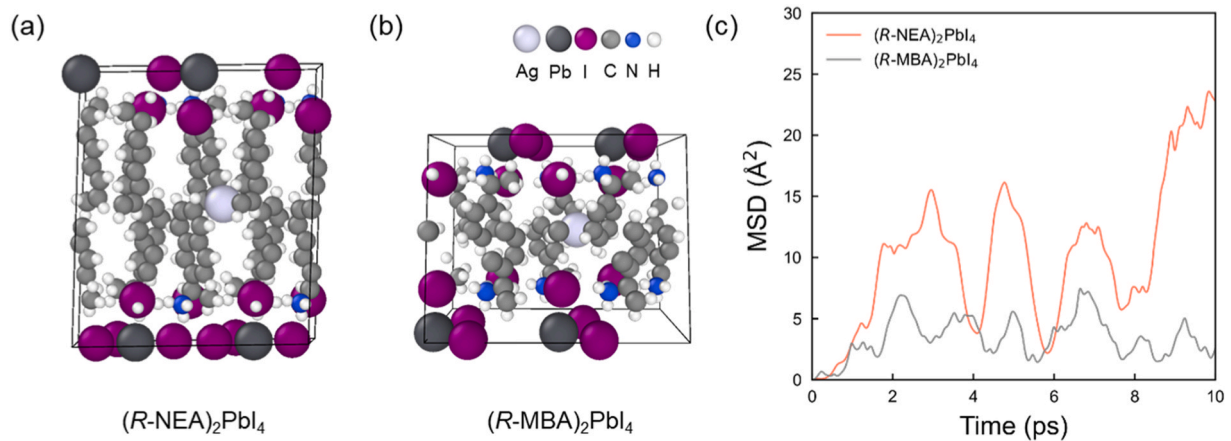


Fig. 5. Atomic structures used in the AIMD simulations for (a) (R-NEA)₂PbI₄ and (b) (R-MBA)₂PbI₄ with an incorporated Ag ion. (c) Mean square displacement (MSD) of an Ag ion in (R-NEA)₂PbI₄ and (R-MBA)₂PbI₄.

of tuning the memory behavior from short-term plasticity (STP) to long-term plasticity (LTP). This controllability could represent an advantage of our platform and suggests a viable strategy for overcoming the intrinsically short retention typically observed in CBRAM devices. Furthermore, by directly correlating the Ag distribution with the retention characteristics, these additional results provide more rigorous experimental evidence supporting the proposed switching mechanism.

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2.6. Chiral synaptic properties of the chiral RRAM

The synaptic functionality of the chiral RRAM was further investigated to evaluate its potential for neuromorphic computing applications. Memristive devices serve as effective representations of artificial synapses as their conductance can be gradually tuned by electrical stimuli, enabling the emulation of synaptic plasticity. In this regard, the CPL-dependent modulation of the HRS current enables multiple conductance states, providing a basis for analog synaptic weight control and potentiation behavior. This behavior allows synaptic potentiation analogous to biological systems. Furthermore, the CPVK exhibits CPL-dependent optoelectronic responses. As shown in Fig. 6a, the device exhibits potentiation characteristics under dark, RCP, and LCP conditions, indicating polarization-dependent synaptic behavior. The electrical pulse measurements (Fig. S17) show that the read current gradually increases with increasing pulse width, indicating a progressive

enhancement of the synaptic weight, which is consistent with the typical behavior of pulse-driven artificial synapses. Furthermore, optical pulse measurements under right- and left-circularly polarized illumination (Fig. S18) reveal a clear polarization-dependent current response, confirming that optical pulses, in addition to electrical pulses, can be utilized to modulate the synaptic characteristics of the proposed device. Although the DC I–V measurements show approximately one order of magnitude difference among the three conditions, the difference becomes less pronounced in pulse-driven synaptic operation. A previous study, a distinct CPL-dependent response was observed only when a sufficiently large gate voltage was applied, indicating that CPL-induced modulation could be effectively manifested only under an adequate electric field [24]. Similarly, in the present device, the relatively thick (~600 nm) perovskite active layer likely requires a sufficiently large bias and/or pulse duration for the CPL-induced modulation to fully influence carrier transport and resistive switching. Consequently, short electrical pulses may not provide sufficient time or driving force for the CPL-induced differences to significantly affect the switching behavior. We anticipate that reducing the active layer thickness and optimizing both carrier and ion transport through device engineering will substantially shorten the required pulse duration while preserving the CPL modulation effect. More importantly, the electric field remains the primary driving force for conductive filament formation, whereas CPL serves as an additional physical control parameter that modulates the electrically driven switching process through spin-polarized carrier generation and spin-selective carrier transport. To evaluate the neuromorphic computing capability of the device, artificial neural network (ANN) simulations were performed using the experimentally obtained

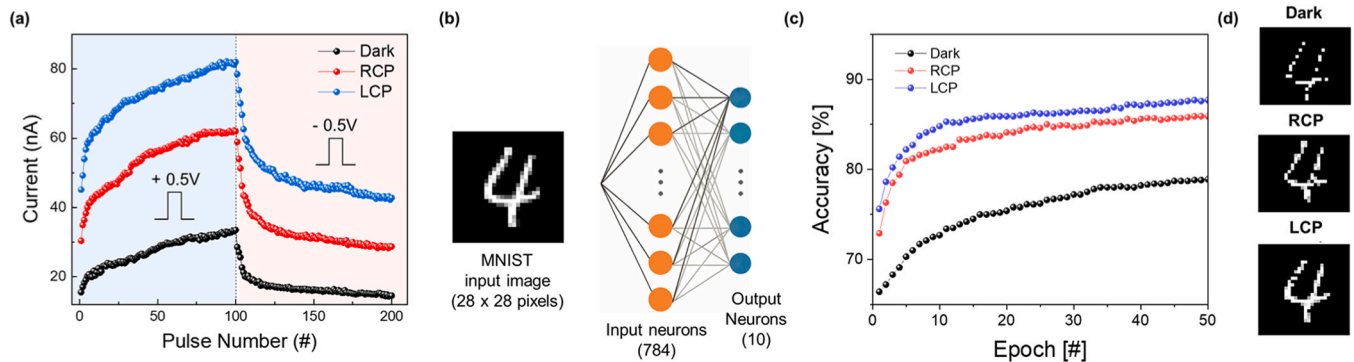


Fig. 6. (a) Potentiation and depression characteristics of the chiral RRAM under dark, RCP, and LCP conditions. (b) Schematic of a single-layer neural networks consisting of 784 input neurons and 10 output neurons. (c) Recognition accuracy for MNIST input images under dark, RCP, and LCP conditions. (d) Reconstructed images after 50 training epochs under dark, RCP and LCP conditions.

potentiation characteristics (Fig. 6b). We employed a single-layer ANN for MNIST handwritten digit recognition, consisting of 784 input neurons and 10 output neurons fully connected through 7840 synapses. Each synaptic weight was modeled using a differential pair of conductance to reflect the device characteristics. Input pixels were pre-processed to emulate the CPL-dependent photo-response, and the network was trained using backpropagation with a sigmoid activation function. The detailed procedures for the state-dependent simulation and learning algorithms are outlined in the Methods Section. As shown in Fig. 6c, the recognition accuracy increases with training epochs and depends on the illumination condition. At 50 training epochs, the recognition accuracy reaches 76% under dark conditions, 83% under RCP illumination, and 87% under LCP illumination, suggesting that CPL-controlled synaptic modulation can improve learning performance. A detailed comparison with chiral perovskite memristive systems is provided in Table S3 to contextualize its performance within the field. Furthermore, the reconstructed images shown in Fig. 6d exhibit clearer patterns under CPL illumination. Because higher conductance states correspond to larger synaptic weights, the increased conductance under CPL conditions leads to stronger pixel activation and clearer reconstructed images. These results highlight the potential of CPL-controlled conductance modulation for neuromorphic applications, demonstrating a viable route toward multi-level memory operation in RRAM-based memristive systems.

3. Conclusion

We demonstrated polarization-controlled resistive switching in a CPVK RRAM device. Light polarization serves as an independent control parameter, enabling deterministic modulation of switching behavior without relying on light intensity. Conduction mechanism analysis reveals that polarization-dependent carrier generation induces a transition from interface-limited to bulk-dominated transport, providing a physical origin for the observed switching modulation. In parallel, ligand-engineered expansion of the interlayer spacing facilitates uniform Ag infiltration, reducing ion migration barriers and enabling homogeneous filament formation, which leads to reduced V_{set} and significantly improved switching uniformity. By combining polarization-dependent electronic control with intrinsic structural design, this approach enables low-voltage, uniform, and multi-level switching without the need for complex engineering strategies. Furthermore, the device exhibits synaptic functionality, and ANN simulations demonstrate enhanced learning performance under CPL illumination. Overall, this work establishes light polarization as a new degree of freedom for controlling resistive switching behavior and provides a simple yet effective strategy for multifunctional memory and neuromorphic systems.

CRediT authorship contribution statement

Kyeounghak Kim: Writing – review & editing. **Young-Hoon Kim:** Writing – review & editing. **Seung-heon Chris Baek:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Conceptualization. **In-Kook Hwang:** Writing – original draft, Formal analysis, Conceptualization. **Cheong Beom Lee:** Writing – original draft, Formal analysis, Data curation. **Si-Yeol Lee:** Methodology. **Sang Hyun Nam:** Methodology, Formal analysis, Writing – review & editing. **Seung-Hwan Kim:** Methodology. **Ji-Hyeon Yun:** Methodology. **Soeun Ahn:** Methodology, Formal analysis, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.nanoen.2026.112278.

Data availability

Data will be made available on request.

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