Hysteretic Ion Migration and Remanent Field in Metal Halide Perovskites

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Abstract: The gap in understanding how underlying chemical dynamics impact the functionality of metal halide perovskites (MHPs) leads to the controversy about the origin of many phenomena associated with ion migration in MHPs. In particular, the debate regarding the impact of ion migration on current–voltage ($I$–$V$) hysteresis of MHPs devices has lasted for many years, where the difficulty lies in directly uncovering the chemical dynamics, as well as identifying and separating the impact of specific ions. In this work, using a newly developed time-resolved time-of-flight secondary ion mass spectrometry $\text{CH}_3\text{NH}_3^+$ and $\text{I}^-$ migrations in $\text{CH}_3\text{NH}_3\text{PbI}_3$ are directly observed, revealing hysteretic $\text{CH}_3\text{NH}_3^+$ and $\text{I}^-$ migrations. Additionally, hysteretic $\text{CH}_3\text{NH}_3^+$ migration is illumination-dependent. Correlating these results with the $I$–$V$ characterization, this work uncovers that $\text{CH}_3\text{NH}_3^+$ redistribution can induce a remanent field leading to a spontaneous current in the device. It unveils that the $\text{CH}_3\text{NH}_3^+$ migration is responsible for the illumination-associated $I$–$V$ hysteresis in MHPs. Hysteretic ion migration has not been uncovered and the contribution of any ions (e.g., $\text{CH}_3\text{NH}_3^+$) has not been specified before. Such insightful and detailed information has up to now been missing, which is critical to improving MHPs photovoltaic performance and developing MHPs-based memristors and synaptic devices.

Metal halide perovskites (MHPs), as a class of outstanding optoelectronic materials,[1] have attracted tremendous research interests in the past decade.[2] MHPs show lots of interesting phenomena when interacting with light, such as phase segregation,[3,4] self-poling,[5] structural transformations,[6] lattice expansion,[7] giant dielectric constant,[8] bulk and interfacial polarization,[9,10] and ultimately photovoltaic performance.[11] However, intrinsic properties related to these phenomena remain elusive. Although many illumination-dependent phenomena in MHPs are proposed to originate from the redistribution of mobile ions and defects, we are still far from fully understanding how illumination affects chemical dynamics and associated physical phenomena in MHPs because direct evidence of illumination-dependent chemical dynamics is rare to date.

MHP is an intricate ionic system containing multiple highly mobile ionic species that can affect the material properties.[1,2,3,12] For instance, both intrinsic (e.g., organic cations and halide anions[13–18] and extrinsic (e.g., sodium, lithium, silver, gold, etc.[19,20]) mobile ions—that can annihilate defects,[21] cause degradation[16,17,19]—have been discovered in MHPs. This leads to superimposed mechanisms of chemophysical interaction in MHPs. However, ion migration in MHPs is usually investigated by indirect methods or studied in static regime, which monitor the physical responses induced by chemical changes or static chemical distribution, thus losing the precise nature of the underlying chemical dynamics. Additionally, in indirect methods, it is challenging to know which mobile species are responsible for the modification of properties because the physical responses can be induced by any ions. These result in longstanding debate about the impact of chemical dynamics on the functionalities of MHPs. A classical example is how chemical dynamics affect the current–voltage ($I$–$V$) hysteresis in MHP solar cells. Ion migration[22] and associated behavior (such as band bending,[23] dynamic trapping/detrapping,[24] and slow transient capacitive current[25,26]) have been proposed to interpret $I$–$V$ hysteresis, as well as the effect of light-soaking and electric poling on current–voltage hysteresis.[27] Nonetheless, a definitive answer to the contribution of ion migration is still lacking. This is due to a lack in experimental evidence that directly provides information regarding ion migration and identifies the contribution of specific ions to the physical properties.
In this work, we directly observe hysteretic CH$_3$NH$_3$$^+$ and I$^-$ migrations and identify the contribution of illumination-dependent CH$_3$NH$_3$$^+$ migration to the $I$–$V$ hysteresis observed in our lateral MHP device, which also implies the potential role of CH$_3$NH$_3$$^+$ in the $I$–$V$ hysteresis in other MHP-based devices. Using an in-house developed time-resolved time-of-flight secondary ion mass spectrometry (tr-ToF-SIMS), we investigated the time evolution of ion distribution in MHPs during the application of external fields, such as external electric field and light illumination. The tr-ToF-SIMS allows us to directly observe the real-time dynamics of any ions (such as CH$_3$NH$_3$$^+$ and I$^-$) in MHPs. Furthermore, taking advantage of machine learning approaches, i.e., nonnegative matrix factorization (NMF), we reveal that both CH$_3$NH$_3$$^+$ and I$^-$ migrations are hysteretic. The CH$_3$NH$_3$$^+$ migration hysteresis is more illumination-dependent, which shows larger loops under light-on condition. In addition, using electric current measurement, we elucidate the effect of ion migration on the dynamic current behavior. We reveal that CH$_3$NH$_3$$^+$ migration can result in a remanent field in the lateral device after the removal of electric bias, which leads to a spontaneous current under illumination. This current remains stable after 200 s of the removal of electric bias, suggesting the remanent field can last >200 s. More importantly, the plot of this spontaneous current as a function of preceding poling bias also shows a hysteresis. We expect this new understanding about the role of mobile ionic species in the hysteretic behavior of MHPs-based device can advance MHPs photovoltaics and stimulate new research interest of MHPs.

To gain deeper insight into the CH$_3$NH$_3$$^+$ migration driven by electric bias, we applied machine learning analysis method of NMF to spatially isolate the tr-ToF-SIMS CH$_3$NH$_3$$^+$ distribution profiles. A more detailed description of NMF analysis is in the Experimental Section. NMF allows us to separate complicated processes or signals into straightforward endmembers that are more physically interpretable. Using NMF the tr-ToF-SIMS result (Figure S3b, Supporting Information) was decomposed into five endmembers (Figure S5a–e, Supporting Information), each endmember describes the CH$_3$NH$_3$$^+$ distributed in the respective regions, where we find an endmember (Figure S5c, Supporting Information) that well represents the CH$_3$NH$_3$$^+$ distributed at Au/CH$_3$NH$_3$PbI$_3$ interfaces (which we named as endmember interface). Here, we emphasize on understanding ionic behavior at Au/CH$_3$NH$_3$PbI$_3$ interfaces as ionic behaviors at interface are critical to device performance, which can modulate the carriers transport barriers, built-in electric field, charge accumulation, etc. Nonetheless, the time evolution of each NMF endmember can be seen in Figure S5f–j (Supporting Information), which shows the periodic cycling during the application of electric bias (Figure S3a, Supporting Information) as expected. In addition, there is an endmember (Figure S5b, Supporting Information) that can describe the CH$_3$NH$_3$$^+$ on electrodes, which we attributed to the contamination on electrodes during the measurement; as shown in Figure S5g (Supporting Information), this endmember is zero initially, suggesting no contamination on the electrodes at the beginning of the measurement, consistent with our expectation. We also tried to isolate the tr-ToF-SIMS result into 2–4 endmembers, but we found that five endmembers are optimal—which simultaneously separate the interested information and exclude the artifacts.
Figure 2. Hysteretic ion migration. a) Voltage–time (V–t) curves of the gradually increasing pulsed stepwise bias that is applied to induce ion migration; b–f) the evolution of interface CH$_3$NH$_3^+$ density as a function of voltage under light-on and light-off conditions; g–k) the evolution of interface I$^-$ density as a function of voltage under light-on and light-off conditions.

We plotted the off-field evolution of the interface endmember as a function of preceding voltage (Figure S6a, Supporting Information) in order to understand how the electric poling modifies the interfacial chemistry in the Au/CH$_3$NH$_3$PbI$_3$/Au device at ground state. Here, each step is 9 s (Figure 2a) in this measurement, so the off-field response showing the dependence of ion distribution on preceding voltages (Figure S6, Supporting Information) suggests that CH$_3$NH$_3^+$ migration can last at least 9 s after the removal of the electric bias. Detailed behaviors of the evolution of the endmember interface during each subperiod are shown separately in Figure 2b–f, where the hysteretic behavior is clearly seen in Figure 2c–f (no hysteretic behavior in Figure 2b). This indicates hysteretic CH$_3$NH$_3^+$ migration can only happen in our device when a bipolar electric waveform with amplitude equal to or greater than 2 V (corresponding to an equivalent average electric field of 80 kV m$^{-1}$) is applied. In addition, the CH$_3$NH$_3^+$ migration hysteresis loops are similar when the bias amplitude increases from 2 to 5 V, as shown in
During electric bias poling, erasure seconds according to the results in Figure 2. Therefore, this charge accumulates at Au/CH$_3$NH$_3$PbI$_3$ interfaces to compensate the polarisation induced by the external electric field, resulting in a charging current; after removal of electric bias, the dissipation of accumulated charges induces a discharging current with opposite sign. We checked further if the current decay observed here is consistent with the capacitive effect decay by fitting them exponentially using a modified capacitive current equation

$$I = I_0 e^{-\frac{t}{RC}} + I_1$$

where $I_0$ is the starting current, $RC$ together specifies the rate of charge/discharge of a capacitor (which is named as RC time constant), where $R$ is the resistance of the circuit and $C$ is the capacity of the device. Considering CH$_3$NH$_3$PbI$_3$ is a semiconductor, we also introduced a stable current $I_s$ to represent the current due to the semiconducting nature of CH$_3$NH$_3$PbI$_3$. As expected, both charging current and discharging current decay exponentially with time (Figure S10a–d, Supporting Information), consistent with capacitive current decay. We also obtained the RC time constant of this device under various voltages (Figure S12, Supporting Information), which increases with the amplitude of the increasing voltage. This is because that the bias poling induces a remanent field persisted after the removal of the bias (the remanent field will be discussed later); as a consequence, the device condition is slightly different at each step. Nonetheless, all the RC time constants are similar (in the range of 17.12–22.09 ms). It is worth noting that we expect faster decay (smaller RC time constant) with the bias voltage increases if this decay process is dominated by ion migration as the ions are expected to move faster under larger bias voltages; therefore, the trend of RC time constant (Figure S12, Supporting Information) also suggests this fast decay process is not dominated by ion migration.

The $I$–$t$ curve under light-on condition was also studied, as shown in Figure S13a (Supporting Information). Similar to that under light-off, light-on electric current also shows a sharp increase in electric current (Figure S13b, Supporting Information) upon the application of electric bias (on-field), which is followed by a fast decay lasting tens to hundreds of milliseconds. After the removal of electric bias (off-field), a sharp spike in current with opposite sign is observed (Figure S13c, Supporting Information), followed by a fast decay lasting tens to hundreds of milliseconds. These decays can also be fitted exponentially (Figure S14, Supporting Information). In addition, the RC time constant also increases with increasing bias voltages (Figure S15, Supporting Information). Therefore, we believe the dominant origin of these fast decays (on-field decays and off-field decays) is still the capacitive effect. However, when we scrutinized the on-field current and the off-field current under light-on condition, we found that the on-field current slowly increases after the initial fast decay (as shown in the inset of Figure S13b, Supporting Information) and the off-field current slowly decreases after the initial fast decay (as shown in the inset of Figure S13c, Supporting Information). In tr-ToF-SIMS ion migration studies, we observed ion migration can last several seconds, consistent with the slow behavior observed in the insets of Figure S13b,c (Supporting Information). Accordingly, we attribute the slow increase of on-field current and the slow decrease of off-field current to the ion migration. In addition, these slow behaviors are not observed under light-off condition, as shown in the insets of Figure S10c,d (Supporting Information), suggesting these slow behaviors are governed...
Figure 3. Current–time (I–t) curves after electric poling. a) The off-field I–t curve under light-on condition along with the V–t curve. b) The plot of the spontaneous current under light-on condition as a function of the on-field bias. c) The off-field I–t curve under light-off condition along with the V–t curve. d) The plot of the spontaneous current under light-off condition as a function of the on-field bias.

by the ion whose behavior is illumination-dependent, that is, \( \text{CH}_3\text{NH}_3^+ \) ion.

Another feature we observed in I–t curve under light-on condition is that the off-field current does not reach 0 A after 9 s relaxation, as shown in the inset of Figure S13c (Supporting Information) (we refer this current to spontaneous current hereafter). This motivated us to explore whether this spontaneous current depends on the preceding bias (i.e., on-field bias). Therefore, we only plotted the off-field I–t curves under light-on condition to highlight the correlation between the spontaneous current and the on-field bias, as shown in Figure 3a. It is clearly seen that the spontaneous current depends on the on-field bias. In the first-half cycle (from 0 to 162 s, in Figure 3a), where the on-field biases are positive, the spontaneous current is negative, suggesting the positive poling generates a negative remanent field that induces a negative spontaneous current after the removal of the bias. When plotting the spontaneous current as a function of the preceding bias, we observed a hysteresis loop, as shown in Figure 3b. Similar analyses were also applied on the I–t curve under light-off condition, as shown in Figure 3c,d. However, all related behaviors are much weaker, or even invisible under light-off. In particular, the hysteresis in Figure 3d is almost closed.

After revealing the hysteretic spontaneous current, we wonder if this spontaneous current is just a temporal current originating from the slow ion diffusion. Therefore, we performed an additional I–t measurement to examine how long the spontaneous current can last. As shown in Figure 4a, we applied 5 V bias to the lateral device for 9 s as for other experiments, then left the device to relax under 0 V for longer time—200 s. The behavior of the electric current during bias poling is the same to that we observed in Figure 3a. When we zoomed in (the inset of the Figure 4a), we can still observe the current around 218 s, suggesting that the spontaneous current can last >200 s. Even if we only measured the current for 200 s after the removal of the electric field, we speculate this spontaneous current can persist permanently because it is still very stable after 200 s. We also performed this measurement by applying 3 V bias to the device, as shown in Figure 4b. The overall phenomena are similar to that by 5 V poling in Figure 4a. In addition, the spontaneous current after 3 V poling (the inset of Figure 4b) is smaller than that after 5 V poling (Figure 4a) as expected. This measurement was also performed under light-off condition as a comparison. As shown in Figure S16 (Supporting Information), both the currents after 5 V poling (Figure S16a, Supporting Information) and 3 V poling (Figure S16b, Supporting Information) are almost zero. This is also consistent with our observation in Figure 3c.

According to these observations, we propose that the \( \text{CH}_3\text{NH}_3^+ \) redistribution partially persists after the removal of the bias voltage, which induces a permanent remanent field leading to a spontaneous current in the device for following reasons. From the electric current measurements, we clearly see
difference of the device condition after bias poling between light-on and light-off. Although we attributed the initial fast decay to capacitive effect, the decay rate should depend on the device condition. The observation of hysteretic behavior suggests that the device condition is different after various bias poling; therefore, the spread of the decay rate (RC time constant) under various bias voltages can be an indicator of how much the device condition is modified by bias poling. As seen in Figures S12 and S15 (Supporting Information), the spread of the RC time constant is Δ = 4.97 ms (from 17.12 to 22.09 ms) under light-off, while it is Δ = 6.81 ms (from 17.89 to 24.70 ms) under light-on, suggesting that the device is modified more significantly under light-on. I− migration is the same under light-on and light-off (Figure 2f), which should not be responsible for these changes. In contrast, CH3NH3+ migration depends on light condition (Figure 2f), implying it is a reason for the device condition change. Moreover, the similarity between the spontaneous current hysteresis and CH3NH3+ migration hysteresis depending on light illumination—both CH3NH3+ migration and spontaneous current show larger hysteresis under light-on—is a more straightforward implication for the correlation between the CH3NH3+ migration and the spontaneous current. Nonetheless, we would like to note that the observation in tr-ToF-SIMS is ionic specific, while the spontaneous current. Nonetheless, we would like to note that the observation in tr-ToF-SIMS is ionic specific, while the spontaneous current behavior. In this work, NMF data analysis was performed on a desk computer using Python 3.6 and scikit-learn 0.19.2 library, where the CH3NH3+ migration plays an indispensable role.

In summary, we for the first time directly observed hysteretic CH3NH3+ and I− migrations using an in-house developed tr-ToF-SIMS technique. We found that CH3NH3+ migration hysteresis is more illumination-dependent than I− migration. We revealed that the bias-induced ion migration under illumination can result in a remanent current lasting more than 200 s, while this remanent current is negligible under light-off. Moreover, the plot of this remanent current under illumination as a function of the bias voltage shows a current–voltage hysteresis loop. Combining the hysteretic ion migration with current–voltage characterization, we revealed that CH3NH3+ migration contributes to the illumination-dependent current–voltage hysteresis. According to the light-dependent behavior of CH3NH3+ migration, we propose that the CH3NH3+ migration also play critical roles in other illumination-dependent phenomena in MHPs. These results offer new and insightful information regarding the understanding of MHPs functionality, which will advance the development of MHPs photovoltaics and stimulate research interests of other MHPs-based functional devices, such as memristors and synaptic devices. In addition, the demonstrated technique, tr-ToF-SIMS, is also useful for other material systems; we expect this method will also be helpful to investigate the chemical changes in other materials.

**Experimental Section**

**CH3NH3PbI3 Synthesis and Lateral Device Fabrication:** The CH3NH3PbI3 samples were synthesized on Swiss glass slides. The glass slides were precleaned using deionized water, acetone, and isopropanol, respectively. The CH3NH3PbI3 was synthesized by spin-coating the precursor (0.8 m lead (II) acetate trihydrate and 2.4 m CH3NH3I in dimethylformamide) and annealed at 100 °C for 30 min in a N2-filled glove box. The gold (Au) electrodes were deposited in a clean room using an electron beam evaporator and a mask.

**Time-of-Flight Secondary Ion Mass Spectrometry:** ToF-SIMS measurements were performed using ToF.SIMS5NCS instrument (IONTOF GmbH, Germany). Experiments were carried out in positive ion mode with spectra calibrated using the Na+, CH3NH3+, and Pb+ peaks for CH3NH3+ studies and in negative ion mode for I− studies with spectra calibrated using the I−, Au+, and I2− peaks. A B2+ analysis beam was used to scan a sample area with 128 × 128 px random pattern. Illumination condition was changed by a built-in LED white light in ToF-SIMS chamber. Bias voltages were applied externally using a Tektronix AFG1022 arbitrary function generator (Tektronix, Beaverton, OR) and Lab VIEW. The current-time (I−t) curve measurements were also conducted in the ToF-SIMS chamber by applying electric waveform using Lab VIEW.

**Nonnegative Matrix Factorization:** NMF is a classical form of the unsupervised unmixing machine learning approach, which is an algorithm in multivariate analysis where an input data is factorized into a number of endmember-abundance pairs with the property that all endmembers have no negative elements. This method has been widely utilized in materials science in analyzing experimental data. This method is particularly suitable for analyzing ToF-SIMS data, where the nonnegativity is inherent because molecular mass cannot be negative.

In this work, NMF data analysis was performed on a desk computer using Python 3.6 and scikit-learn 0.19.2 library, where the CH3NH3+ distribution profile Xj is unmixed on a linear combination of endmembers vn and noise Nj.

\[
X_j = \sum A_i v_i + N_j
\]
endmembers are calculated by minimizing noise $N$, component, subject to $v_i \geq 0$, which means every endmember will be nonnegative. More information regarding NMF and its application in material science can be found elsewhere.[3]

**Statistical Analysis**: Experiments and data analysis details were demonstrated above in each subsection of the Experimental Section. The current-time results are shown as raw data and smoothed data, the smoothed data are obtained by smoothing the raw data with Savitzky–Golay filter in Python.

**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.

**Keywords**

hysteresis, ion migration, metal halide perovskites, remanent fields, spontaneous polarization

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