Breakthrough architecture conquers dark current: echoes of Dujiangyan in perovskite photodetectors

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Metal halide perovskites (MHPs) photodetectors (PDs) exhibiting outstanding and tunable photoelectric properties have attracted extensive global attention owing to their low-temperature solution-processable fabrication, lightweight nature, and flexibility.[1] However, MHPs-based PDs still face challenges of high and unstable dark current, which limit their operational stability and practical applications.[2] Minimizing dark current is crucial for enhancing the sensitivity of detectors, determining the limit of detection (LoD) of PDs. In photoconductive PDs, reducing defect density in semiconductor materials to enhance resistivity is a common technique for reducing dark current.[3] Alternatively, interface barriers, such as those in metal-semiconductor-metal (MSM) structures, can prevent charge injection and substantially decrease dark current.[4] However, because both photocurrent and dark current are collected through the same electrode pathway, these strategies to lower dark current often result in reduced photocurrent as well, thereby limiting improvements in sensitivity. Furthermore, the phenomenon of dark current drift (also known as baseline drift or unstable dark current) in perovskite PDs, a noticeable yet often overlooked concern, has consistently hindered its commercialization.[5] Dark current drift leads to challenges in accurately assessing device photodetection performance and significantly diminishes operational reliability, particularly under low-light conditions. Therefore, there is a pressing demand to thoroughly investigate the mechanism of dark current generation and drift in perovskite detectors and to propose viable solutions.

Now, writing in an issue of *Nature Communications*, Bowen Zhu's team from Westlake University clarified the ion migration as the primary cause of unstable dark current in perovskite PDs, and provided a novel device structure to simultaneously solve the challenges of high and unstable dark current in photoconductor-type perovskite PDs.[6]

Organic cations [such as $CH_3NH_3^+$ (MA^+)] and halide anions (such as I^-) within MHPs exhibit a counter-directional movement along the electric field. This phenomenon, termed ion migration, arises from the weak ionic bonding characteristic of perovskite structures. Remarkably, ion migration has been unequivocally identified as the primary contributor to the hysteresis phenomenon observed in perovskite solar cells.[7] Ion migration can also result in the deterioration of the perovskite structure, thereby compromising the performance of perovskite devices and significantly impacting their operational stability.[8] Moreover, ion migration could potentially be the culprit behind the phenomenon of dark current drift in MHPs-based PDs. However, few studies have been able to provide compelling evidence, either directly or indirectly, to substantiate the aforementioned speculations.

As illustrated in Fig. 1(a), Tang et al. proposed that in conventional photoconductor-type MHPs-based PDs (device#1), when an external bias is applied, the MA^+ ions and I^- ions separate along the direction of the electric field (E), accumulating at the two ends to form an ionic electric field (E_{ionic}) opposing the E. Due to the gradual strengthening of the E_{ionic} over time, the dark current decreases with time, giving rise to the phenomenon of dark current drift [green line in Fig. 1(c)]. Therefore, if it were possible to introduce a uniform distribution of potential in the device, it could effectively suppress ion migration caused by the electric field. In their work, the researchers added a control electrode at the bottom of the conventional photoconductor device to create a longitudinal electric field, ensuring a more uniform distribution of potential [Fig. 1(b)]. The research results indicate that the electrical modulation introduced by the three-electrode structure indeed effectively restrains the dark current drift in perovskite PDs. This indirectly verifies that ion migration serves as the primary culprit behind the occurrence of dark current drift in perovskite PDs. Furthermore, the authors compared the photoluminescence (PL) spectra of the traditional two-electrode device with the newly designed three-electrode device before and after approximately one hour of operation. The results demonstrated that the new structure effectively maintains a more stable PL spectrum compared to the conventional device. This directly verifies the effectiveness of this new structure in inhibiting ion migration, consequently solidifying ion migration as the primary cause behind dark current drift in perovskite PDs. What's even more pleasantly surprising is that this three-electrode structure ingeniously provides distinct transmission pathways for both dark current and photocurrent. The modulation electric field guides the dark current toward the control electrode, effectively enabling the device to detect a signal current with nearly imperceptible dark current. Thereby, owing to the remarkable contributions of the breakthrough architecture, an exceptionally high-performance perovskite PD with ultra-low dark current of 5 pA and an almost imperceptible current drift ($\sim 1.3 \times 10^{-5}$ pA per second) has been successfully developed.

As we know, most previous researchers focused on enhancing the performance and stability of perovskite optoelectronic devices by addressing perovskite defect states and grain boundaries, aiming to block ion migration pathways. The approach presented in this article embodies the wisdom of ancient Chinese water management techniques. By incorporating a control electrode, a fresh architectural paradigm emerges in perovskite PDs, reminiscent of the Dujiangyan system. The Dujiangyan system addressed the concerns surrounding flood disasters and sediment accumulation by constructing diversion levees [Fig. 1(d)]. Similarly, the newly designed perovskite photodetector, featuring a control electrode, skillfully addresses the challenges arising from high dark current and device instability caused by ion migration. This novel structure holds significant implications for suppressing dark current in other materials-based photoconductor-type PDs as well. In addition, this novel detector structure endowing ultra-low and ultra-stable dark current has significant implications for advancing the development of perovskite-based single-pixel imaging technology.[9–11] The next crucial step involves investigating whether this structure can also play a vital role in perovskite photodiode-type PDs.

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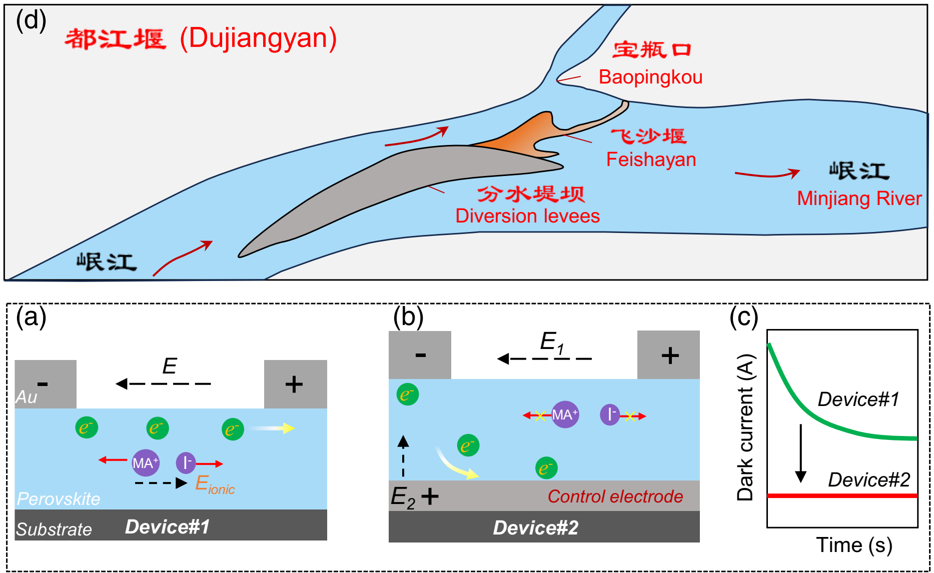

Fig. 1 Echoes of Dujiangyan in perovskite PDs. (a) Working mechanism of conventional photoconductive-type perovskite PDs (device#1). In this device, the signal electrode collects both dark current and photocurrent, increasing low signal-to-noise ratio. Additionally, the operating voltage accelerates ion migration, causing dark current drift. (b) Working mechanism of new structure perovskite PDs (device#2). In this device, a control electrode separates dark current and photocurrent, boosting the signal-to-noise ratio. Moreover, it ensures uniform potential distribution, curbing ion migration and improving dark current drift. The function of the control electrode bears a striking resemblance to the diversion levees in the Dujiangyan. (c) Dark current curves of device#1 and device#2. (d) Schematic diagram of the Dujiangyan Irrigation System. Ancient Chinese ingeniously managed flood discharge and sediment removal along the Minjiang River by constructing engineering works like diversion levees, sediment-flushing weirs (Feishayan), and bottleneck channels (Baopingkou).

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