Solution-Processed Mixed-Dimensional Hybrid Perovskite/Carbon Nanotube Electronics

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Abstract

Benefiting from their extraordinary physical properties, methylammonium lead halide perovskites (PVKs) have attracted significant attention in optoelectronics. However, the PVK-based devices suffer from low carrier mobility and high operation voltage. Here, we utilize sorted semiconducting single-walled carbon nanotubes (95% s-SWCNT) to enhance the performance of thin-film transistors (TFTs) based on the mixed-cation perovskite $(\text{MA}_{1-x}\text{FA}_x)\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$, enabling mixed-dimensional solution-processed electronics with high mobility ($32.25 \text{ cm}^2/\text{Vs}$) and low voltage ($\sim 3 \text{ V}$) operation. The resulting mixed-dimensional PVK/SWCNT TFTs possess ON/OFF ratios on the order of 10^7 , enabling the fabrication of high-gain inverters.

Keywords: perovskite, SWCNT, TFT, electronics, mixed-dimensional vdW heterostructure

In addition to their well-known application in photovoltaic devices (certified power conversion efficiencies exceeding 23%,¹ PVKs have been explored for other devices such as photodetectors,²⁻⁵ light-emitting devices,⁶ and field-effect transistors.⁷⁻¹⁰ In particular, light-emitting diodes based on organometallic halide PVKs have shown a high external quantum efficiency of 20.7% with a power conversion efficiency of 12%.⁶ PVK-based photodetectors have also exhibited excellent performance including high sensitivities and wide spectral responses ranging from ultraviolet-visible-near-infrared (UV-NIR) ³ to X rays ¹¹⁻¹³ and gamma rays.^{14,15} In PVKs, strong inter-band transitions¹⁶ are accompanied by dominant bimolecular recombination below the Langevin limit ^{17,18} and low trap densities.^{19,20} These characteristics, combined with the

photo-recycling effect,^{21,22} give rise to high light absorption and long carrier diffusion lengths,²³ suggesting a high potential for solution-processed PVK optoelectronics.

PVKs are also expected to accelerate the development of solution-processed electronics. **Table S1** provides a list of representative studies on PVK-based thin-film transistors (TFTs). In one of the first studies, Mitzi *et al.* reported a (PEA)₂SnI₄ TFT with a mobility of 0.6 cm²/Vs.²⁴ More recent work has focused on MAPbI_{3-x}Cl_x TFTs with reported mobilities in the range of 1.01-1.24 cm²/Vs.¹⁰ Several efforts have attempted to improve TFT performance by optimizing PVK composition,²⁵ fabricating hybrid bilayers,^{26,27} and using single-crystal microplates.⁹ A particularly powerful approach is to interface PVK with high-mobility nanomaterials such as graphene ^{28,29} and carbon nanotubes (CNTs).⁷ However, these devices have thus far suffered from low ON/OFF ratios and high threshold voltages,^{30,31} limiting their electronic applications.

Single-walled carbon nanotubes (SWCNTs) are favorable for solution-processed TFT applications because of their electronic tunability,³² high carrier mobility,³³ exceptional thermal conductivity and mechanical properties,³⁴ and inherent dispersibility in organic solvents compatible with PVK precursor solutions.³⁵ With high mechanical resilience, SWCNTs have also sparked great interest for stretchable and flexible electronics.³⁶ Most SWCNT synthetic methods result in a 1:2 ratio of metallic (m-SWCNTs) to semiconducting (s-SWCNTs) species.^{37,38} Unlike m-SWCNTs that have no bandgap,³⁹ s-SWCNTs have a tunable bandgap and can form type-II heterostructures with PVKs.^{40,41} Previously, our group reported mixed-dimensional heterostructures based on 3D PVKs and unsorted 1D SWCNTs,⁴² but these devices suffered from high OFF currents (10 mA at 5 V) and low ON/OFF ratios of 10², which is a result of residual m-SWCNTs in the mixture.⁴³ Isolation and enrichment of s-SWCNTs can be accomplished by several methods such as gel-permeation chromatography,⁴⁴ density gradient ultracentrifugation (DGU),⁴⁵

and selective dispersion with conjugated polymers.⁴⁶ However, high-purity s-SWCNTs have not yet been utilized in PVK TFTs, suggesting an opportunity for improved performance. In such a mixed-dimensional heterostructure,⁴⁷ the 1D s-SWCNTs play a key role of transporting the carriers transferred from the 3D PVK matrix. Both SWCNT and PVK are promising to be applied in other mixed-dimensional van der Waals (vdW) heterostructures where materials with different dimensions adhere primarily through non-covalent interactions.⁴⁷

Here, the DGU technique is used to enrich polychiral s-SWCNTs produced by the high pressure carbon monoxide (HiPCo) growth method, allowing the production of TFTs using mixed-cation PVKs ($\text{MA}_{1-x}\text{FA}_x$) $\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$ and sorted s-SWCNTs. Compared to previous devices based on unsorted SWCNTs, the OFF currents of TFTs with sorted s-SWCNTs are significantly suppressed by 4 orders of magnitude, resulting in record-high ON/OFF ratios (10^7). Furthermore, we employed a high- κ gate dielectric layer (HfO_2) to dramatically reduce the TFT threshold voltage from ~ 60 V (SiO_2 devices) to ~ 3 V, enabling low-voltage operation. A thin self-assembled monolayer (SAM) of ethoxylated polyethyleneimine (PEIE) was further used to enhance device stability. To demonstrate the utility of these TFTs for digital electronics, we fabricated a solution-processed inverter with desirable characteristics based on PVKs and s-SWCNTs. As a mixed-dimensional heterostructure, the PVK/s-SWCNT composite exhibits unique physical phenomena such as a transition from high-temperature ambipolar to low-temperature unipolar charge transport behavior, which is a result of competing carrier trapping and emission.

Result and Discussion

Both PVKs and SWCNTs were prepared in solution. The inset in **Figure 1** presents a photograph of the PVK precursor solutions with different concentrations of SWCNTs (pristine, 0.1 wt.%, 0.3 wt.%, and 0.5 wt.%). As our previous work demonstrated,⁷ the PVK precursor

solution of polar, non-hydrogen-bonding Lewis base solvents, N,N-dimethylformamide (DMF) and dimethyl sulfoxide (DMSO), allows for the stable, homogeneous dispersion of SWCNTs.⁴⁰ We used a two-step, anti-solvent engineering method to deposit the PVK films,^{48,49} which is illustrated in **Figure 1a**. Different from our previous work,⁷ here we used precursors of MAI, FABr, PbI₂, and PbBr₂ mixed in DMF/DMSO (4:1) to prepare mixed-cation and mixed-halide (MA_{1-x}FA_x)Pb(I_{1-x}Br_x)₃ perovskites. Thin films based on mixed-cation and mixed-halide PVKs were found to be more stable and possess higher carrier mobility than the commonly used MAPbI₃ perovskite.^{50,51} The improved properties of the mixed-cation PVKs could be a result of their cubic structure with a tolerance factor close to 1. After spin-coating and annealing at 100 °C for 1 hour, dense and uniform polycrystalline PVK thin films embedded with SWCNTs were achieved. More experimental details are given in the experimental section.

Figure 1. Schematic of fabrication and characterization of PVK/SWCNT transistors. a) Photograph of the PVK precursor solutions with different concentrations of SWCNTs and schematic illustration of the two-step solvent engineering process for the fabrication of films composed of mixed-cation PVK and SWCNTs. b) Top-view SEM image of a typical PVK/s-SWCNT film with a SWCNT concentration of 0.1 mg/mL. c) AFM image of the as-prepared PVK/SWCNT film. d) Device structure of the PVK/SWCNT phototransistor, accompanied by schematic representations of the PVK/SWCNT channel and the molecular PEIE self-assembled layer on the top of Au electrodes. The channel length (L) is 50 μm, and the channel width (W) is 1 mm. e) Band alignment of the mixed-dimensional PVK/s-SWCNT channel.

Figures 1b, c show the top-view scanning electron microscopy (SEM) and atomic force microscopy (AFM) images, respectively, of the as-prepared PVK/SWCNT thin film. The SWCNTs appear to be uniformly distributed in the PVK thin film with no evidence of agglomerates or bundles. The solvent molecules and ionic species in the PVK precursor solutions likely interact with the SWCNTs through van der Waals and Coulombic/steric forces that act to

reduce aggregation and promote uniform dispersion.⁵² Furthermore, the PVK/SWCNT thin film showed continuous surface coverage without pinholes.

The bottom-gate, bottom-contact PVK/SWCNT TFT is illustrated in **Figure 1d**. HfO₂ with a thickness of 50 nm deposited by atomic layer deposition (ALD) is used as the gate dielectric layer, and the heavily *n*-doped Si substrate is used as the back gate.⁵³ The relative permittivity (ϵ) of the HfO₂ dielectric was characterized to be 19 (**Figure S1** (d)), which is substantially higher than that of SiO₂ ($\epsilon=3.7$). Au electrodes with a thickness of 80 nm were sputtered on the HfO₂ layer. The channel length between electrodes was 50 μ m, and the channel width was 1 mm. The thin HfO₂ layer with high permittivity affords the low-bias operation of the hybrid PVK/SWCNT electronic devices.

As illustrated in **Figure 1d**, after the electrode deposition, a thin (approximately 2.5 nm) self-assembled monolayer (SAM) of ethoxylated polyethyleneimine (PEIE) was deposited on the substrate. The morphology and thickness of PEIE were characterized by atomic force microscope (AFM) and ellipsometry (**Figure S2**). This layer provides several benefits in terms of device stability and performance. First, it protects the Au electrodes from reacting directly with the PVK layer. As demonstrated in **Figure S3**, without the protective PEIE layer, the Au electrode was partially etched. X-ray photoelectron spectroscopy (XPS) taken on the Au surface without the PEIE protection (**Figure S4**) show a blue-shift of the Au 4f_{7/2} and 4f_{5/2} peaks,⁵⁴ providing additional evidence of reaction between the Au surface and the perovskite layer. Second, the self-assembled dipoles in the thin PEIE layer likely improve the quality of the PVK layer by passivation of the interface.⁵⁵ Third, the PEIE layer facilitates the PVK growth and enhances the substrate adhesion of the PVK grains (**Figure S3**). Finally, from an energetic point of view, PEIE increases

the Fermi level of Au by 0.1 eV, thereby facilitating charge injection by lowering the height of the Schottky barrier between the Au electrode and the semiconductor thin film.⁸

The mixed-dimensional PVK/SWCNT heterostructure is illustrated in the inset of **Figure 1d**. XPS spectra in **Figure S5** confirm the existence of SWCNTs in the PVK film. **Figure 1e** schematically shows the electronic band alignment between the SWCNTs and PVK in the channel layer, together with the Au electrodes and PEIE interlayer. Despite the insulation nature of PEIE, its inherent dipoles and ultrathin nature allow electrons to inject efficiently into the semiconductor channel *via* tunneling and thermionic injection. Because of the type-II band alignment, exciton separation is expected at the PVK/SWCNT interface due to the built-in potential. The separated transport of electrons and holes is beneficial to enhancing the charge diffusion by reducing Coulomb scattering and recombination.⁴⁰

We first characterized the SWCNT samples using optical absorption. **Figure 2a** shows the optical absorbance spectra of the metallic-enriched, semiconducting-enriched, and unsorted SWCNTs. Absorbance of both sorted semiconducting and unsorted dispersions is suppressed in the wavelength range from 440 to 600 nm, the region associated with the first-order excitations (M11) of the SWCNT metallic species. Further analysis revealed that the sorted s-SWCNTs consist of approximately 95% semiconducting species, while the unsorted s-SWCNTs consist of approximately 80% semiconducting species. Overall, these results are consistent with previous reports on semiconducting enrichment of HiPCo SWCNTs by DGU.⁴⁵

Figure 2. Optical and structural characterizations. a) UV-Vis spectra of metallic-enriched, semiconducting-enriched, and unsorted SWCNTs. M11 is the peak associated with the SWCNT metallic species, and S22/S33 represent the SWCNT semiconducting species. b) XRD patterns of the PVK/SWCNT films with different types of SWCNTs (95% semiconducting, 80% semiconducting, and metallic SWCNTs). c) UV-Vis absorption spectra of the PVK/SWCNT films with different types of SWCNTs. d) Corresponding photoluminescence spectra of the different PVK/SWCNT films.

To further investigate the structure of the PVK/SWCNT film, X-ray diffraction (XRD) was performed. **Figure 2b** shows the XRD patterns of the PVK films with different types of SWCNTs with 0.02 w.t.% concentration. Peaks at 14.21° , 28.51° , 43.82° can be assigned to the (110), (220), (330) crystallographic planes of $(\text{MA}_{1-x}\text{FA}_x)\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$, and the peak at 13.94° belongs to residual

PbI₂, which may help passivate the PVK grains.⁵⁶ Interestingly, under the same measurement conditions, the X-ray diffraction peaks of the sorted PVK/s-SWCNT films were sharper and stronger than those of PVK/m-SWCNT films. A zoom-in comparison of the (110) XRD peaks of three samples is presented in **Figure S6**. Our result indicates that the uniformity of sorted s-SWCNTs enhances the crystallinity of the PVK films, which is consistent with the larger grain size observed in the SEM images (**Figure S7**). Because most structural defects exist at the grain boundaries,⁵⁷ we expect reduced charge scattering and improved transport properties in the PVK films with sorted s-SWCNTs.

The structural modification of the PVK films by the s-SWCNTs also impacts the optical properties of the hybrid films. **Figure 2c** shows the UV-vis absorption spectra of PVK films with different types of SWCNTs. All films exhibit an absorption edge at 770 nm, which is consistent with the PVK bandgap.^{25,50} The optical absorption in the PVK/SWCNT films is enhanced in the short wavelength range (300-500 nm) compared to the pristine PVK film. This enhancement in absorption is expected because the addition of SWCNTs improves the morphology and crystallinity of the PVK film.⁴⁰

As shown in **Figure 2d**, substantial photoluminescence (PL) quenching was observed in the hybrid PVK/SWCNT film, confirming the formation of a type-II heterojunction with efficient exciton dissociation at the interfaces. The most significant PL quenching occurs in the hybrid PVK film embedded with enriched s-SWCNTs. Besides the PL quenching, a blue shift (25 nm or 53 meV) of the PL peak is observed with increasing purity of s-SWCNTs, implying the suppression of in-gap trap states.¹⁹

We fabricated bottom-contact, bottom-gate PVK/SWCNT TFTs with Au electrodes and a HfO₂ gate dielectric layer. After testing dozens of devices, the thickness of the PVK film was optimized to be 140 nm (**Figure S8**). If the channel film was too thin, it was discontinuous with poor morphology. On the other hand, if the film was too thick, the gate bias effect was reduced due to screening. **Figure 3a** is the transfer curve (I_{DS} vs. V_g) of the TFT with a pristine PVK channel. We swept the back-gate voltage V_g at a rate of 0.5 V/s, while maintaining the source-drain voltage V_{DS} at a fixed bias (-1 V). The pristine PVK TFT shows a clear ambipolar behavior with both electron and hole transport, which is consistent with previous reports.^{7,58} However, the source-drain current (I_{DS}) is lower than 0.1 μ A, implying overall poor charge transport for pristine PVK. The hole mobility (μ) was extracted using the following relationship:⁷

$$I_{DS} = \mu C_i \frac{W}{L} \left((V_g - V_{TH}) V_{DS} - \frac{V_{DS}^2}{2} \right) \quad (1)$$

where C_i is gate capacitance (355.6 nF/cm²). The hole mobility of the pristine PVK film was calculated to be 0.033 cm²/Vs.

The transport characteristics of the PVK/SWCNT TFTs sensitively depend on the SWCNT concentration. **Figures 3b** and **3c** show the transfer curves of PVK films with s-SWCNTs of 0.1 mg/mL (0.02 wt.%, PVK+SWCNT-01) and 0.5 mg/mL (0.1 wt.%, PVK+SWCNT-05), respectively. The source-drain current increases dramatically with increasing SWCNT concentration, achieving a high hole mobility of 18.3 cm²/Vs at an unsorted s-SWCNT concentration of 0.02 wt.%. The transfer curve of PVK+SWCNT-01 remains ambipolar with suppressed hysteresis, which implies suppressed trap states and improved sample quality after the addition of high-purity SWCNTs. The high-mobility SWCNTs embedded in the PVK matrix

provide pathways for hole transport, and the efficient charge separation reduces electron-hole recombination at grain boundaries. In contrast, the PVK+SWCNT-05 device showed a *p*-type transfer curve with a high OFF current. Although the hole mobility is still high (8.9 cm²/Vs), it suffers from a relatively low ON/OFF ratio (less than one order of magnitude). At this high concentration, the SWCNT percolation threshold is exceeded, resulting in transport being dominated by *p*-type SWCNTs in addition to increased screening that suppresses the ON/OFF ratio. The performance of s-SWCNT TFTs were also investigated. As shown in **Figure S9(a)**, normal spin-coating only results in aggregation of SWCNT clusters. To compare with the s-SWCNT networks in the composites, we used the suction filtration method to prepare homogeneous SWCNT networks shown in **Figure S9(b)**. The transfer curve of the pristine s-SWCNT exhibits *p*-type characteristics and a gate tunability of three orders of magnitude (**Figure S9(c)**).

Figure 3d,e,f show the top-view SEM images of the PVK films with different concentrations of SWCNTs. Faceted grains were observed in the pristine film (**Figure 3d**), and they become larger when SWCNTs with a concentration of 0.1 mg/mL were added to the samples (**Figure 3e**). The high crystallinity of the PVK/SWCNT film benefit both the optical absorption (**Figure 2c**) and the charge transport (**Figure 3b**). On the other hand, as shown in **Figure 3f**, at the highest SWCNT concentration (0.5 mg/mL), large SWCNT bundles are observed, which explains the high OFF current in the TFT. In order to determine the effect of SWCNT type, we compared the performance of TFTs with metallic-enriched, semiconducting-enriched, and unsorted SWCNTs. **Figure 3g** shows the transfer curve of the PVK/m-SWCNT device, which is highly conductive with negligible gate tuning. In contrast, improved performance was observed in TFTs with unsorted (**Figure 3h**) and semiconducting-enriched (**Figure 3i**) SWCNTs. In particular, high-

purity s-SWCNTs maintain the high ON current while suppressing the OFF current to a low level (10^{-10} A). The device with high-purity s-SWCNTs exhibits a hole mobility of $32.2 \text{ cm}^2/\text{Vs}$ in linear region. Furthermore, an ON/OFF ratio of 10^7 was observed.

Figure 3. Performance of PVK/SWCNT TFTs. Transfer characteristics of (a) the pristine mixed-cation PVK TFT and the devices with (b) 0.1 mg/mL 80% s-SWCNTs and (c) 0.5 mg/mL 80% s-SWCNTs. (d-f) Corresponding top-view SEM images of the PVK films with different concentrations of SWCNTs. The scale bars are $1 \mu\text{m}$. (g) Transfer curve of the PVK/m-SWCNTs transistor with a metallic-enriched SWCNT concentration of 0.05 mg/mL. (h,i) transfer curves of the PVK/SWCNT TFTs with 0.1 mg/mL of unsorted and semiconducting-enriched (95%) SWCNTs, respectively.

It should be noted that all the transport measurements above were carried out in the dark and vacuum. Under light illumination, the off-current increases because of photo-induced carriers, leading to a decreased ON/OFF ratio. Nevertheless, as **Figure S10** shows, with the increase of off-current, the PVK/SWCNT TFT can operate as a phototransistor with a gain of above 200 and an ON/OFF ratio of 10^6 .

Figure 4. PVK/SWCNT inverter. a) Transfer curve and b) output curve of the *p*-type dominant PVK/SWCNT TFT with V_{DS} of -1 V. c) Transfer curve and d) output curve of the *n*-type dominant PVK/SWCNT TFT with V_{DS} of 5 V. e) Schematic illustration of the PVK/SWCNT inverter, consisting of two identical PVK/SWCNT TFTs (s -SWCNT concentration: 0.1 mg/mL). Also shown is an optical microscopy image of a TFT channel. f) Input-output curve of the PVK/SWCNT inverters with drive voltages at 5 V. The dashed lines show the voltage-gain characteristics.

To illustrate the utility of the PVK/SWCNT TFTs in digital electronics, a PMOS inverter was fabricated and characterized as shown in **Figure 4**. Specifically, **Figures 4a** and **4b** show the transfer and the output curves, respectively, of the *p*-type dominant PVK/SWCNT TFT. The output curves exhibit saturation at ~ 5 V and a clockwise hysteresis, which may result from the trap states in the PVK film.⁸ **Figures 4c** and **4d** show the transfer curve and the output curve of the *n*-type dominant PVK/SWCNT TFT with V_{DS} at 5 V. **Figure 4e** illustrates the schematic of the PVK/SWCNT inverter. The inverter was connected in a complementary configuration,²⁵ consisting of two identical PVK/SWCNT TFTs. **Figure 4f** shows the voltage transfer characteristics (V_{OUT} vs V_{IN}) of the inverters with different supply voltages V_{DD} . At ~ 1 V, the input signal was inverted with sharp switching. As shown in **Figure 4f**, when the supply voltage is 5 V, the gain of the inverter can achieve the maximum value of 7.6. The trip voltage (~ 1 V) is close to the ideal trip voltage ($V_{DD}/2$), which leads to the stable operation of the inverters. Compared to previous work,⁸ the use of the HfO₂ dielectric layer lowers the threshold voltage below 3 V, making such PVK/SWCNT devices promising for low-voltage optoelectronics.

To gain insight on the carrier transport and trapping, we measured the TFT characteristics at variable temperatures (**Figures 5a** and **5b**). Four conductance regions of the PVK/SWCNT channel can be defined. As temperature increases, the transition from region (I) to (II) can be attributed to the phase transition (from tetragonal to orthorhombic) occurring at around 160 K in PVK films, which was reported to affect charge transport.⁵⁹ When the temperature further increases to room temperature, the conductance starts to decrease from region (II) to (III). One possible reason behind this transition is that ions defreeze at high temperature, and the ion screening and scattering effects limit the charge transport. Finally, region (IV) shows an emergence

of electron conductance at high positive bias, and electrons that are trapped at low temperatures start to be released from ionized traps when the temperature increases above 160 K.⁶⁰

Figure 5. Temperature-dependent performance of the PVK/SWCNT TFT. Transfer curves of the PVK/SWCNT TFT measured at different temperatures with a) forward scanning and b) reverse scanning. c) Hole mobility and trap density extracted from the temperature-dependent transfer curves. Insets are the schematic of the crystal structures of perovskites at low temperature (orthorhombic) and room temperature (tetragonal). d) Conductance of the PVK/SWCNT channel at different temperatures.

As shown in **Figure S11a**, at a low temperature of 80 K, the transfer curve of the transistor shows an anti-clockwise hysteresis, and the p -type behavior becomes much pronounced. With temperature increasing to 120 K, the ON/OFF ratio slightly decreases while the p -type behavior is

sustained (**Figure S11b**). At 160 K, the hysteresis loop significantly shrinks, indicating the decrease of effective trap density (**Figure S11c**). When the temperature continues increasing to 200 K, the shape of the hysteresis loop changes as a result of the emergence of *n*-type transport (**Figure S11d**). At such intermediate temperatures, electrons are involved in the carrier transport, although their role is still moderate compared to holes. This trend continues at 240 K (**Figure S11e**), and at 280 K the PVK/SWCNT channel shows ambipolar characteristics. As a general trend, with increasing temperature, the hysteresis loop continues shrinking, indicating the suppression of the effects of both electron and hole traps.

Since in the sub-threshold region channel transport can be tuned by the gate voltage without saturation, the PVK/SWCNT film is in the flat-band region and less affected by space charges.⁶¹ In this region, the hole mobility can be extracted from

$$I_{DS} = \mu_{Linear} C_i \frac{W}{L} \left((V_g - V_{TH}) V_{DS} - \frac{V_{DS}^2}{2} \right) \quad (2)$$

$$\mu_{Linear} = \frac{L}{C_i W V_{DS}} \frac{\partial I_{DS}}{\partial V_g}. \quad (3)$$

The linear fittings are given in **Figure S12**. As shown in **Figure 5c**, the hole mobility decreases with increasing temperature, which is in line with the general behavior of semiconductors. The hole mobility of the transistor is calculated to be ~80 cm²/Vs at 80 K. At higher temperatures, both phonon scattering and ion migration contribute to the scattering cross-section of holes. However, a plateau of hole mobility is observed at ~160 K, which coincides with the structural transition of perovskites as reported in previous studies.^{8,59} We conducted a temperature dependent PL measurement on the mixed-dimensional SWCNT/PVK heterostructure in **Figure S13**. We

observed the disappearance of the low-temperature PL peak at ~160 K, confirming the phase transition. Therefore, we conclude the origin of the change of conductance at ~160 K is related to the phase transition of the PVK. Furthermore, the total trap density can be determined by the hysteresis loop in the transfer curves, *i.e.*, $N_{\text{TT}} = C_{\text{ox}}\Delta V_{\text{TH}}/q$, in which $C_{\text{ox}} = \epsilon_{\text{ox}}/t_{\text{ox}}$ is the oxide capacitance, q is the elementary charge, and ΔV_{TH} is the difference of V_{TH} when scanning in the forward and reverse directions.⁶⁰ As shown in **Figure 5c**, the effective trap density decreases with increasing temperature, which results from the higher kinetic energy of carriers and higher emission probability. The trap density is $7.3 \times 10^{12} \text{ cm}^{-2}$ at 80 K and decreases to $8.8 \times 10^{11} \text{ cm}^{-2}$ at room temperature, which is quite close to the values reported for pure MAPbI₃ films,⁶² indicating that SWCNTs do not exacerbate defect trapping in PVK films. The trap density is close to that of typical vdW heterostructures such as MoS₂/hBN,⁶³ so that the hysteresis of transfer characteristic is quite moderate at room temperature. However, we observed large hysteresis in the output curve shown in **Figure 4**, which is likely a result of ion migration and bulk impurities.

Figure 5d shows the temperature-dependent conductivity (σ) of the PVK/SWCNT transistor channel at $V_g = 2 \text{ V}$ and $V_{\text{DS}} = -1 \text{ V}$. The conductance was calculated by $\sigma = L/tWR$, in which L is the channel length (50 μm), W is the channel width (1000 μm), and t is the film thickness (140 nm). We observed a clear transition of conductance at 160 K. Since ion migration is suppressed at low temperatures and TFTs show *p*-type behavior when the temperature is below 160 K, we can assume holes as the dominant carriers. From fitting to the Arrhenius plot in **Figure S14**, we obtained the activation energy (E_a) of holes as 99.1 meV. First-principles calculation by Yin *et al.* predicted that the electronic activation energy of MA vacancies V_{MA} is as small as 50 meV.⁶⁴ Temperature-dependent transient response measurements performed by Li *et al.* obtained an activation energy of $120 \pm 10 \text{ meV}$.⁶⁵ Four-probe Hall and SCLC measurements on perovskite

single crystals reported by Adinolfi *et al.* revealed a hole trapping state peak at 0.1 eV and an electron trapping state peak at 0.2 eV.¹⁹ The experimentally obtained hole activation energy is approximately twice the theoretical prediction, which indicates additional charge trapping caused by disorder introduced in solution processing. Furthermore, the fact that the activation energy obtained here is comparable to previous reports on pure perovskite films implies that the incorporation of s-SWCNTs does not exacerbate the carrier trapping. Finally, **Figure 5d** shows that when the temperature is above 160 K, the conductance of films decreases with increasing temperature, which indicates enhanced charge scattering caused by ion migration at high temperatures.

Figure 6. Schematics of trapping-induced hysteresis and ambipolar-to-unipolar transition.

a) Schematics of the transfer curves at low temperature and room temperature. Insets are schematics of dominant carriers trapping in perovskite trap states, which generates hysteresis. At room temperature, the increased carrier velocity eliminates the effective trapping states. b) Trapping (*p*-type trapping state) of mobile carriers close to or in the channel (dominant at low temperatures) causes the hysteresis; above the phase transition temperature (160 K) of the perovskite film, the transistor behavior changes from *p*-type to ambipolar. At the same time, trapping is weaker at higher temperatures, which suppresses hysteresis.

The temperature-dependent TFT transfer characteristics provide valuable insights into the mechanism of carrier transport in the PVK/SWCNT heterostructure. As shown in the schematics in **Figure 6a** (data in **Figure S11**), there are two key features of the transfer curves: one is that the channel shows *p*-type behavior at low temperature and ambipolar behavior at room temperature; the other is that the hysteresis at low temperature is much more pronounced than that at room temperature. These observations suggest that the temperature-dependent characteristics of charge traps play a key role in channel transport.

According to the SRH (Shockley-Read-Hall) model (**Figure 6b**), carriers from the valence band/conduction band are captured by trap states at a rate proportional to the capture cross-section multiplied by the thermal velocity of the carriers, or $P_{capture} \propto \sqrt{T}$.⁶⁶ In addition, based on the Heuristic interpretation of the SRH model, considering the different occupation probabilities of the energy levels, the probability of emission is raised by the Boltzmann factor, or $P_{emission} \propto \sqrt{T} \exp\left(-\frac{E_a}{kT}\right)$. As a result, charge capture dominates at low temperatures, but the probability of emission increases faster with increasing temperature.⁶⁰ The *p*-type transfer curves at low temperatures indicate that electrons are captured by trap states and the channel transport is dominated by holes. However, more electrons are released from traps with increasing temperature, leading to the ambipolar nature of perovskite transistors at room temperature. Our data imply that hole trap states are located sufficiently close to the valence band edge so that holes can be effectively emitted into the valence band at low temperatures, leading to the observed *p*-type transport. In contrast, since electron trap states are located deeper in the bandgap, negligible concentrations of electrons are released to the conduction band at low temperatures. At room

temperature, the population of trap states are reduced because of the higher kinetic energy of electrons and higher probability of emission, which leads to the ambipolar behavior.

Conclusion

In summary, semiconducting-enriched single-walled carbon nanotubes (95% s-SWCNT) have been used to enhance the performance of mixed-cation perovskite transistors and inverters, enabling solution-processed mixed-dimensional electronics with high mobility and low operation bias. The mixed-dimensional PVK/SWCNT heterostructures exhibited a low OFF current (10^{-11} A), leading to an ultrahigh ON/OFF ratio up to 10^7 . Furthermore, we fabricated high-gain inverters with a switching voltage of 1 V, which provides opportunities for digital electronic applications. Finally, our temperature-dependent transport study revealed a transition from high-temperature ambipolar to low-temperature unipolar behavior, which is a result of competing carrier trapping and emission. The demonstrated merits of low-cost solution processing, high carrier mobility, low-voltage operation, high ON/OFF ratio, and versatile operation modes make mixed-dimensional PVK/SWCNT heterostructures a highly promising candidate for a variety of electronic applications.

Experimental Section

SWCNT film preparation

Unsorted (80% semiconducting purity) SWCNTs were dispersed by sonicating 75 mg HiPCO (NanoIntegris) powder, 10 wt% SWCNTs and 90 wt% binder, in 15 ml 1% w/v sodium

dodecyl sulfate in deionized water by a 0.125 inch tip horn ultrasonicator at 13 W for 60 min, cooled by an ice water bath. The resulting solution was then centrifuged at 10,400g for 15 minutes, to separate the individual SWCNTs in the supernatant from the SWCNT bundles, carbonaceous impurities, and metal catalyst particles in the pellet. The supernatant was vacuum filtered on a Nylon membrane and rinsed with copious amounts of ethanol, acetone, and isopropanol to remove residual surfactant. Films were then air dried, peeled off the nylon membrane, and placed in vials for later processing. For sorted (95% semiconducting purity) semiconducting single-walled carbon nanotubes (s-SWCNTs), 150 mg HiPCo powder was similarly dispersed in an aqueous solution of 1% w/v sodium cholate and 25.6% w/v iodixanol for the same power and duration as the unsorted solution. After centrifuging the sonicated solutions at 10,400g for 15 min, the supernatant was subsequently sorted by electronic type using density gradient ultracentrifugation and characterized for electronic-type purity as previously reported.⁴⁵ Purified fractions were combined into one solution, which was then vacuum filtered and stored as stated above.

PVK/SWCNT precursor preparation

The SWCNTs were dissolved in DMF (Sigma-Aldrich, 99.8%) and DMSO (Sigma-Aldrich, 99.7%) solvent (DMF/DMSO = 4:1 volume ratio) after which the dispersion solvent was ultrasonicated for 8 hours and then added to the mixed compounds. The perovskite precursor solution contains 0.8 M Methylammonium iodide (MAI, Dyesol), 0.8 M lead (II) iodide (PbI₂, Sigma-Aldrich, 98%), 0.08 M formamidinium bromide (FABr, Dyesol), and 0.08 M lead (II) bromide (PbBr₂, Sigma-Aldrich, 98%). The mixed precursor was ultra-sonicated for 2 hours before deposition.

Fabrication and characterization of TFT devices

Heavily doped Si substrates were cleaned with hydrofluoric acid (HF) and ultrasonicated in deionized water, ethanol, and acetone, respectively. After drying in flowing nitrogen, a thin layer (50 nm) of HfO₂ was grown by atomic layer deposition. The Si/HfO₂ substrates were treated with oxygen plasma before film deposition. Au (80 nm) source (S) and drain (D) electrodes were deposited by thermal evaporation through a shadow mask, defining a channel with length of 50 μm and width of 1000 μm . A PEIE solution (0.4 w.t.% in 2-methoxyethanol) was spin coated on the electrodes at 5000 rpm for 30 s. The substrates were then heated at 100 °C for 10 minutes, and followed with ultrasonication in DI water for 10 minutes. A thin layer of PEIE (~5 nm) was deposited after drying the substrates in nitrogen and annealing at 100 °C for 5 minutes in a glovebox. The mixed solution was then spin-coated on the substrates at 2000 rpm for 10 s. A two-step spin coating was performed at 6000 rpm for 30 s, followed by 50 μL of anti-solvent (toluene) dripping during the last 10 s, after which the devices were annealed at 100 °C for 1 hour in a glove box to reduce the charge traps and improve the Ohmic contact between the active layer and electrodes. For comparison, the pristine perovskite films were prepared without SWCNTs following the same protocol.

XRD was performed on a Bruker D8-advanced diffractometer using Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$). The morphology of the films was measured using a FEI Nova Nano 630, and AFM images were obtained on a Bruker Icon AFM. PL measurements were carried out using a Horiba Aramis Raman system. Absorption spectra were obtained using an Agilent Cary 5000 UV–vis–NIR spectrometer. Temperature-dependent transport curves were measured using a Keithley 4200 Semiconductor Parametric Analyzer and Lakeshore CPX cryogenic probe station in dark and vacuum, where the voltage sweep rate was 0.5 V/s.

Supporting Information

The Supporting Information is available free of charge online at

Characterization of the HfO₂ and the measurement setup (Figure S1); characterization of the PEIE layer (Figure S2); images of the Au electrodes w, w/t PEIE protection (Figure S3); XPS spectra of Au electrodes w, w/t PEIE protection (Figure S4); XPS spectra of PVK films w, w/t SWCNT (Figure S5); Enlarged XRD patterns of PVK with different SWCNTs (Figure S6); SEM images of PVKs with different SWCNTs (Figure S7); profilometry data of the PVK films (Figure S8); SEM images of the SWCNTs prepared by different methods and its transfer curve (Figure S9); Photoresponse of the PVK/SWCNT TFT (Figure S10); temperature-dependent measurements of the PVK/SWCNT TFT (Figure S11); transfer curves of the PVK/SWCNT TFTs (Figure S12); temperature-dependent PL measurement on SWCNT/PVK heterostructure (Figure S13); fitting of activation energy (Figure S14); A comparison of studies of transistors and inverters based on PVKs (Table S1).

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References

1. Jeon, N. J.; Na, H.; Jung, E. H.; Yang, T. Y.; Lee, Y. G.; Kim, G.; Shin, H. W.; Seok, S. I.; Lee, J.; Seo, J., A Fluorene-Terminated Hole-Transporting Material for Highly Efficient and Stable Perovskite Solar Cells. *Nat. Energy* **2018**, 3, 682-689.

2. Wang, H.; Kim, D. H., Perovskite-Based Photodetectors: Materials and Devices. *Chem. Soc. Rev.* **2017**, *46*, 5204-5236.
3. Dou, L.; Yang, Y. M.; You, J.; Hong, Z.; Chang, W. H.; Li, G.; Yang, Y., Solution-Processed Hybrid Perovskite Photodetectors with High Detectivity. *Nat. Commun.* **2014**, *5*, 5404.
4. Ma, C.; Shi, Y. M.; Hu, W. J.; Chiu, M. H.; Liu, Z. X.; Bera, A.; Li, F.; Wang, H.; Li, L. J.; Wu, T., Heterostructured WS₂/CH₃NH₃PbI₃ Photoconductors with Suppressed Dark Current and Enhanced Photodetectivity. *Adv. Mater.* **2016**, *28*, 3683-3689.
5. Fang, Y. J.; Huang, J. S., Resolving Weak Light of Sub-Picowatt Per Square Centimeter by Hybrid Perovskite Photodetectors Enabled by Noise Reduction. *Adv. Mater.* **2015**, *27*, 2804-2810.
6. Ling, Y.; Yuan, Z.; Tian, Y.; Wang, X.; Wang, J. C.; Xin, Y.; Hanson, K.; Ma, B.; Gao, H., Bright Light-Emitting Diodes Based on Organometal Halide Perovskite Nanoplatelets. *Adv. Mater.* **2016**, *28*, 305-11.
7. Li, F.; Wang, H.; Kufer, D.; Liang, L. L.; Yu, W. L.; Alarousu, E.; Ma, C.; Li, Y. Y.; Liu, Z. X.; Liu, C. X.; Wei, N. N.; Wang, F.; Chen, L.; Mohammed, O. F.; Fratolocchi, A.; Liu, X. G.; Konstantatos, G.; Wu, T., Ultrahigh Carrier Mobility Achieved in Photoresponsive Hybrid Perovskite Films *via* Coupling with Single-Walled Carbon Nanotubes. *Adv. Mater.* **2017**, *29*, 1602432.
8. Senanayak, S. P.; Yang, B. Y.; Thomas, T. H.; Giesbrecht, N.; Huang, W. C.; Gann, E.; Nair, B.; Goedel, K.; Guha, S.; Moya, X.; McNeill, C. R.; Docampo, P.; Sadhanala, A.; Friend, R. H.; Sirringhaus, H., Understanding Charge Transport in Lead Iodide Perovskite Thin-Film Field-Effect Transistors. *Sci. Adv.* **2017**, *3*, e1601935.
9. Wang, G. M.; Li, D. H.; Cheng, H. C.; Li, Y. J.; Chen, C. Y.; Yin, A. X.; Zhao, Z. P.; Lin, Z. Y.; Wu, H.; He, Q. Y.; Ding, M. N.; Liu, Y.; Huang, Y.; Duan, X. F., Wafer-Scale Growth of Large Arrays of Perovskite Microplate Crystals for Functional Electronics and Optoelectronics. *Sci. Adv.* **2015**, *1*, e1500613.
10. Li, F.; Ma, C.; Wang, H.; Hu, W. J.; Yu, W. L.; Sheikh, A. D.; Wu, T., Ambipolar Solution-Processed Hybrid Perovskite Phototransistors. *Nat. Commun.* **2015**, *6*, 8238.
11. Kim, Y. C.; Kim, K. H.; Son, D. Y.; Jeong, D. N.; Seo, J. Y.; Choi, Y. S.; Han, I. T.; Lee, S. Y.; Park, N. G., Printable Organometallic Perovskite Enables Large-Area, Low-Dose X-Ray Imaging. *Nature* **2017**, *550*, 87-91.
12. Shrestha, S.; Fischer, R.; Matt, G. J.; Feldner, P.; Michel, T.; Osvet, A.; Levchuk, I.; Merle, B.; Golkar, S.; Chen, H. W.; Tedde, S. F.; Schmidt, O.; Hock, R.; Ruhrig, M.; Goken, M.; Heiss, W.; Anton, G.; Brabec, C. J., High-Performance Direct Conversion X-Ray Detectors Based on Sintered Hybrid Lead Triiodide Perovskite Wafers. *Nat. Photonics* **2017**, *11*, 436-440.
13. Palazon, F.; Akkerman, Q. A.; Prato, M.; Manna, L., X-Ray Lithography on Perovskite Nanocrystals Films: From Patterning with Anion-Exchange Reactions to Enhanced Stability in Air and Water. *ACS Nano* **2016**, *10*, 1224-1230.
14. Yakunin, S.; Dirin, D. N.; Shynkarenko, Y.; Morad, V.; Cherniukh, I.; Nazarenko, O.; Kreil, D.; Nauser, T.; Kovalenko, M. V., Detection of Gamma Photons Using Solution-Grown Single Crystals of Hybrid Lead Halide Perovskites. *Nat. Photonics* **2016**, *10*, 585-589.
15. Wei, H. T.; DeSantis, D.; Wei, W.; Deng, Y. H.; Guo, D. Y.; Savenije, T. J.; Cao, L.; Huang, J. S., Dopant Compensation in Alloyed CH₃NH₃PbBr₃-xCl_x Perovskite Single Crystals for Gamma-Ray Spectroscopy. *Nat. Mater.* **2017**, *16*, 826-833.
16. Herz, L. M., Charge-Carrier Mobilities in Metal Halide Perovskites: Fundamental Mechanisms and Limits. *ACS Energy Lett.* **2017**, *2*, 1539-1548.

17. Johnston, M. B.; Herz, L. M., Hybrid Perovskites for Photovoltaics: Charge-Carrier Recombination, Diffusion, and Radiative Efficiencies. *Acc. Chem. Res.* **2016**, *49*, 146-154.
18. Xing, G. C.; Wu, B.; Wu, X. Y.; Li, M. J.; Du, B.; Wei, Q.; Guo, J.; Yeow, E. K. L.; Sum, T. C.; Huang, W., Transcending the Slow Bimolecular Recombination in Lead-Halide Perovskites for Electroluminescence. *Nat. Commun.* **2017**, *8*, 14558.
19. Adinolfi, V.; Yuan, M.; Comin, R.; Thibau, E. S.; Shi, D.; Saidaminov, M. I.; Kanjanaboos, P.; Kopilovic, D.; Hoogland, S.; Lu, Z. H.; Bakr, O. M.; Sargent, E. H., The In-Gap Electronic State Spectrum of Methylammonium Lead Iodide Single-Crystal Perovskites. *Adv. Mater.* **2016**, *28*, 3406-3410.
20. Shi, D.; Adinolfi, V.; Comin, R.; Yuan, M. J.; Alarousu, E.; Buin, A.; Chen, Y.; Hoogland, S.; Rothenberger, A.; Katsiev, K.; Losovyj, Y.; Zhang, X.; Dowben, P. A.; Mohammed, O. F.; Sargent, E. H.; Bakr, O. M., Low Trap-State Density and Long Carrier Diffusion in Organolead Trihalide Perovskite Single Crystals. *Science* **2015**, *347*, 519-522.
21. Richter, J. M.; Abdi-Jalebi, M.; Sadhanala, A.; Tabachnyk, M.; Rivett, J. P. H.; Pazos-Outon, L. M.; Godel, K. C.; Price, M.; Deschler, F.; Friend, R. H., Enhancing Photoluminescence Yields in Lead Halide Perovskites by Photon Recycling and Light Out-Coupling. *Nat. Commun.* **2016**, *7*, 13941.
22. Pazos-Outon, L. M.; Szumilo, M.; Lamboll, R.; Richter, J. M.; Crespo-Quesada, M.; Abdi-Jalebi, M.; Beeson, H. J.; Vrucinic, M.; Alsari, M.; Snaith, H. J.; Ehrler, B.; Friend, R. H.; Deschler, F., Photon Recycling in Lead Iodide Perovskite Solar Cells. *Science* **2016**, *351*, 1430-1433.
23. Stranks, S. D.; Eperon, G. E.; Grancini, G.; Menelaou, C.; Alcocer, M. J. P.; Leijtens, T.; Herz, L. M.; Petrozza, A.; Snaith, H. J., Electron-Hole Diffusion Lengths Exceeding 1 Micrometer in an Organometal Trihalide Perovskite Absorber. *Science* **2013**, *342*, 341-344.
24. Kagan, C. R.; Mitzi, D. B.; Dimitrakopoulos, C. D., Organic-Inorganic Hybrid Materials as Semiconducting Channels in Thin-Film Field-Effect Transistors. *Science* **1999**, *286*, 945-947.
25. Yusoff, A. B.; Kim, H. P.; Li, X.; Kim, J.; Jang, J.; Nazeeruddin, M. K., Ambipolar Triple Cation Perovskite Field Effect Transistors and Inverters. *Adv. Mater.* **2017**, *29*, 1062940.
26. Cheng, H. C.; Wang, G. M.; Li, D. H.; He, Q. Y.; Yin, A. X.; Liu, Y.; Wu, H.; Ding, M. N.; Huang, Y.; Duan, X. F., Van der Waals Heterojunction Devices Based on Organohalide Perovskites and Two-Dimensional Materials. *Nano Lett.* **2016**, *16*, 367-373.
27. Lin, Y. H.; Huang, W. T.; Pattanasattayavong, P.; Lim, J.; Li, R. P.; Sakai, N.; Panidi, J.; Hong, M. J.; Ma, C.; Wei, N. N.; Wehbe, N.; Fei, Z. P.; Heeney, M.; Labram, J. G.; Anthopoulos, T. D.; Snaith, H. J., Deciphering Photocarrier Dynamics for Tuneable High-Performance Perovskite-Organic Semiconductor Heterojunction Phototransistors. *Nat. Commun.* **2019**, *10*, 4475.
28. Lee, Y.; Kwon, J.; Hwang, E.; Ra, C. H.; Yoo, W. J.; Ahn, J. H.; Park, J. H.; Cho, J. H., High-Performance Perovskite-Graphene Hybrid Photodetector. *Adv. Mater.* **2015**, *27*, 41-46.
29. Chang, P. H.; Liu, S. Y.; Lan, Y. B.; Tsai, Y. C.; You, X. Q.; Li, C. S.; Huang, K. Y.; Chou, A. S.; Cheng, T. C.; Wang, J. K.; Wu, C. I., Ultrahigh Responsivity and Detectivity Graphene-Perovskite Hybrid Phototransistors by Sequential Vapor Deposition. *Sci. Rep.* **2017**, *7*, 46281.
30. De Volder, M. F. L.; Tawfick, S. H.; Baughman, R. H.; Hart, A. J., Carbon Nanotubes: Present and Future Commercial Applications. *Science* **2013**, *339*, 535-539.
31. Javey, A.; Guo, J.; Wang, Q.; Lundstrom, M.; Dai, H. J., Ballistic Carbon Nanotube Field-Effect Transistors. *Nature* **2003**, *424*, 654-657.

32. Xie, Y.; Gong, M. G.; Shastry, T. A.; Lohrman, J.; Hersam, M. C.; Ren, S. Q., Broad-Spectral-Response Nanocarbon Bulk-Heterojunction Excitonic Photodetectors. *Adv. Mater.* **2013**, *25*, 3433-3437.
33. Durkop, T.; Getty, S. A.; Cobas, E.; Fuhrer, M. S., Extraordinary Mobility in Semiconducting Carbon Nanotubes. *Nano Lett.* **2004**, *4*, 35-39.
34. Avouris, P.; Freitag, M.; Perebeinos, V., Carbon-Nanotube Photonics and Optoelectronics. *Nat. Photonics* **2008**, *2*, 341-350.
35. Ausman, K. D.; Piner, R.; Lourie, O.; Ruoff, R. S.; Korobov, M., Organic Solvent Dispersions of Single-Walled Carbon Nanotubes: Toward Solutions of Pristine Nanotubes. *J. Phys. Chem. B* **2000**, *104*, 8911-8915.
36. Yamada, T.; Hayamizu, Y.; Yamamoto, Y.; Yomogida, Y.; Izadi-Najafabadi, A.; Futaba, D. N.; Hata, K., A Stretchable Carbon Nanotube Strain Sensor for Human-Motion Detection. *Nat. Nanotechnol.* **2011**, *6*, 296-301.
37. Ghosh, S.; Bachilo, S. M.; Weisman, R. B., Advanced Sorting of Single-Walled Carbon Nanotubes by Nonlinear Density-Gradient Ultracentrifugation. *Nat. Nanotechnol.* **2010**, *5*, 443-450.
38. Baughman, R. H.; Zakhidov, A. A.; de Heer, W. A., Carbon Nanotubes - the Route Toward Applications. *Science* **2002**, *297*, 787-792.
39. Matsuda, Y.; Tahir-Kheli, J.; Goddard, W. A., Definitive Band Gaps for Single-Wall Carbon Nanotubes. *J. Phys. Chem. Lett.* **2010**, *1*, 2946-2950.
40. Schulz, P.; Dowgiallo, A. M.; Yang, M. J.; Zhu, K.; Blackburn, J. L.; Berry, J. J., Charge Transfer Dynamics between Carbon Nanotubes and Hybrid Organic Metal Halide Perovskite Films. *J. Phys. Chem. Lett.* **2016**, *7*, 418-425.
41. Blackburn, J. L., Semiconducting Single-Walled Carbon Nanotubes in Solar Energy Harvesting. *ACS Energy Lett.* **2017**, *2*, 1598-1613.
42. Li, F.; Wang, H.; Kufer, D.; Liang, L. L.; Yu, W. L.; Alarousu, E.; Ma, C.; Li, Y. Y.; Liu, Z. X.; Liu, C. X.; Wei, N. N.; Wang, F.; Chen, L.; Mohammed, O. F.; Fratalocchi, A.; Liu, X. G.; Konstantatos, G.; Wu, T., Ultrahigh Carrier Mobility Achieved in Photoresponsive Hybrid Perovskite Films via Coupling with Single-Walled Carbon Nanotubes. *Adv. Mater.* **2017**, *29*, 1602432.
43. Arnold, M. S.; Green, A. A.; Hulvat, J. F.; Stupp, S. I.; Hersam, M. C., Sorting Carbon Nanotubes by Electronic Structure Using Density Differentiation. *Nat. Nanotechnol.* **2006**, *1*, 60-65.
44. Flavel, B. S.; Moore, K. E.; Pfohl, M.; Kappes, M. M.; Hennrich, F., Separation of Single-Walled Carbon Nanotubes with a Gel Permeation Chromatography System. *ACS Nano* **2014**, *8*, 9687-9687.
45. Green, A. A.; Hersam, M. C., Nearly Single-Chirality Single-Walled Carbon Nanotubes Produced via Orthogonal Iterative Density Gradient Ultracentrifugation. *Adv. Mater.* **2011**, *23*, 2185-2190.
46. Nish, A.; Hwang, J. Y.; Doig, J.; Nicholas, R. J., Highly Selective Dispersion of Single-Walled Carbon Nanotubes Using Aromatic Polymers. *Nat. Nanotechnol.* **2007**, *2*, 640-646.
47. Jariwala, D.; Marks, T. J.; Hersam, M. C., Mixed-Dimensional Van der Waals Heterostructures. *Nat. Mater.* **2017**, *16*, 170-181.
48. Jeon, N. J.; Noh, J. H.; Kim, Y. C.; Yang, W. S.; Ryu, S.; Seok, S. I., Solvent Engineering for High-Performance Inorganic-Organic Hybrid Perovskite Solar Cells. *Nat. Mater.* **2014**, *13*, 897-903.

49. Jeon, N. J.; Noh, J. H.; Yang, W. S.; Kim, Y. C.; Ryu, S.; Seo, J.; Seok, S. I., Compositional Engineering of Perovskite Materials for High-Performance Solar Cells. *Nature* **2015**, *517*, 476-480.
50. Saliba, M.; Matsui, T.; Domanski, K.; Seo, J. Y.; Ummadisingu, A.; Zakeeruddin, S. M.; Correa-Baena, J. P.; Tress, W. R.; Abate, A.; Hagfeldt, A.; Gratzel, M., Incorporation of Rubidium Cations into Perovskite Solar Cells Improves Photovoltaic Performance. *Science* **2016**, *354*, 206-209.
51. Salado, M.; Calio, L.; Berger, R.; Kazim, S.; Ahmad, S., Influence of the Mixed Organic Cation Ratio in Lead Iodide Based Perovskite on the Performance of Solar Cells. *Phys. Chem. Chem. Phys.* **2016**, *18*, 27148-27157.
52. Pramanik, C.; Gissinger, J. R.; Kumar, S.; Heinz, H., Carbon Nanotube Dispersion in Solvents and Polymer Solutions: Mechanisms, Assembly, and Preferences. *ACS Nano* **2017**, *11*, 12805-12816.
53. Wang, H.; Yu, L. L.; Lee, Y. H.; Shi, Y. M.; Hsu, A.; Chin, M. L.; Li, L. J.; Dubey, M.; Kong, J.; Palacios, T., Integrated Circuits Based on Bilayer MoS₂ Transistors. *Nano Lett* **2012**, *12*, 4674-4680.
54. Kundu, S.; Wang, Y. M.; Xia, W.; Muhler, M., Thermal Stability and Reducibility of Oxygen-Containing Functional Groups on Multiwalled Carbon Nanotube Surfaces: A Quantitative High-Resolution XPS and TPD/TPR Study. *J. Phys. Chem. C* **2008**, *112*, 16869-16878.
55. Bai, Y.; Dong, Q. F.; Shao, Y. C.; Deng, Y. H.; Wang, Q.; Shen, L.; Wang, D.; Wei, W.; Huang, J. S., Enhancing Stability and Efficiency of Perovskite Solar Cells with Crosslinkable Silane-Functionalized and Doped Fullerene. *Nat. Commun.* **2016**, *7*, 12806.
56. Chen, Q.; Zhou, H. P.; Song, T. B.; Luo, S.; Hong, Z. R.; Duan, H. S.; Dou, L. T.; Liu, Y. S.; Yang, Y., Controllable Self-Induced Passivation of Hybrid Lead Iodide Perovskites toward High Performance Solar Cells. *Nano Lett.* **2014**, *14*, 4158-4163.
57. Yang, M. J.; Zeng, Y. N.; Li, Z.; Kim, D. H.; Jiang, C. S.; van de Lagemaat, J.; Zhu, K., Do Grain Boundaries Dominate Non-Radiative Recombination in CH₃NH₃PbI₃ Perovskite Thin Films? *Phys. Chem. Chem. Phys.* **2017**, *19*, 5043-5050.
58. Li, D. H.; Cheng, H. C.; Wang, Y. L.; Zhao, Z. P.; Wang, G. M.; Wu, H.; He, Q. Y.; Huang, Y.; Duan, X. F., The Effect of Thermal Annealing on Charge Transport in Organolead Halide Perovskite Microplate Field-Effect Transistors. *Adv. Mater.* **2017**, *29*, 1601959.
59. Wu, K. W.; Bera, A.; Ma, C.; Du, Y. M.; Yang, Y.; Li, L.; Wu, T., Temperature-Dependent Excitonic Photoluminescence of Hybrid Organometal Halide Perovskite Films. *Phys. Chem. Chem. Phys.* **2014**, *16*, 22476-22481.
60. Park, R. S.; Shulaker, M. M.; Hills, G.; Liyanage, L. S.; Lee, S.; Tang, A.; Mitra, S.; Wong, H. S. P., Hysteresis in Carbon Nanotube Transistors: Measurement and Analysis of Trap Density, Energy Level, and Spatial Distribution. *ACS Nano* **2016**, *10*, 4599-4608.
61. Zhang, Z.; Yates, J. T., Band Bending in Semiconductors: Chemical and Physical Consequences at Surfaces and Interfaces. *Chem. Rev.* **2012**, *112*, 5520-5551.
62. Zhang, X. N.; Bi, S. Q.; Zhou, J. Y.; You, S.; Zhou, H. Q.; Zhang, Y.; Tang, Z. Y., Temperature-Dependent Charge Transport in Solution-Processed Perovskite Solar Cells with Tunable Trap Concentration and Charge Recombination. *J. Mater. Chem. C* **2017**, *5*, 9376-9382.
63. Illarionov, Y. Y.; Rzepa, G.; Watzl, M.; Knobloch, T.; Grill, A.; Furchi, M. M.; Mueller, T.; Grasser, T., The Role of Charge Trapping in MoS₂/SiO₂ and MoS₂/hBN Field-Effect Transistors. *2D Mater.* **2016**, *3*, 035004.

64. Yin, W. J.; Shi, T. T.; Yan, Y. F., Unusual Defect Physics in CH₃NH₃PbI₃ Perovskite Solar Cell Absorber. *Appl. Phys. Lett.* **2014**, *104*, 063903.
65. Li, D. H.; Wu, H.; Cheng, H. C.; Wang, G. M.; Huang, Y.; Duan, X. F., Electronic and Ionic Transport Dynamics in Organolead Halide Perovskites. *ACS Nano* **2016**, *10*, 6933-6941.
66. Egginger, M.; Bauer, S.; Schwodiauer, R.; Neugebauer, H.; Sariciftci, N. S., Current *versus* Gate Voltage Hysteresis in Organic Field Effect Transistors. *Monatsh Chem.* **2009**, *140*, 735-750.

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Solution-Processed Mixed-Dimensional Hybrid Perovskite/Carbon Nanotube Electronics

TOC