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Electrical and Optoelectrical Dual-Modulation in Perovskite-Based Vertical Field-Effect Transistors

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ABSTRACT: Organolead trihalide perovskites have drawn a great deal of interest for application in electronic and optoelectronic devices owing to their exceptional physical properties. However, the majority of reported perovskite field-effect transistors (FETs) demonstrates unsatisfactory performance, such as poor mobility, low on/off ratio, and low current density, which remains a significant obstacle for their potential applications. In this work, we report a vertical FET (VFET) composed of a single-crystalline MAPbBr₃ perovskite Schottky junction, which effectively modulates the intrinsic carrier characteristics and enhances the



performance of the device by reducing channel length and weakening the electrostatic screening effect. The device exhibits excellent performance with a high on/off ratio (10^4), high carrier mobility ($23.4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$), and current density of 2.69 A cm⁻² at room temperature. Importantly, the regulation of the drain voltage plays a similar role to that of the gate voltage in our device, which is attributed to the doping of perovskite with regulation of the source contact/perovskite Schottky junction. With the drain modulation, a superior photoresponse with enhanced responsivity of 16 A W^{1–} and a low dark current are simultaneously obtained under light illumination, leading to a high detectivity of 10^{14} Jones. Our research may open up new possibilities for perovskite electronics, such as logic applications for various flexible, wearable, and disposable devices.

KEYWORDS: perovskite microplates, vertical field-effect transistor, high performance, dual modulation

1. INTRODUCTION

Perovskite-structure materials are of great interest in science and semiconductor devices.¹⁻⁴ Among them, the organicinorganic mixed halide perovskites have attracted the most attention in a variety of energy-related and optoelectronic devices because of the remarkable physical and chemical properties, including a tunable bandgap,⁵ extraordinary absorption coefficient,⁶ outstanding carrier mobility,⁷⁻⁹ and long carrier diffusion lengths.¹⁰ Specially, the power conversion efficiency of perovskite solar cells has rapidly increased over a decade, from 3.8% to a recent record value of 25.6%,^{1,11} and the external quantum efficiency of perovskite light-emitting diodes (LEDs) has grown by an order of magnitude in only a few years.¹² In addition to applications in solar cells and LEDs, the research community has also investigated the use of these materials in the transistor technologies, owing to their inherent high carrier mobility (over 100 cm² V⁻¹ s⁻¹ measured by terahertz techniques).^{13–15} Although it has been known that the perovskite-based FETs can provide better performance because of the remarkable intrinsic carrier mobility themselves, the performance of most perovskite-based FETs reported to date is unsatisfactory in comparison to the theoretical predictions. The experimental reported devices always suffer from poor mobilities induced by substantial grain boundaries and serious carrier scattering and trapping, low on/off ratios by the electrostatic screening effect, and long-term operational

instability. Currently, the mobilities obtained based on perovskite FEFs are much lower than optimal values, e.g., a maximum value of around 4.7 cm² V⁻¹ s⁻¹ was observed in MAPbX₃ perovskite FETs¹⁶ and a maximum of around 2 cm² V⁻¹ s⁻¹ in triple cation perovskites.¹⁷ Additionally, large on/off ratios obtained from FETs can only be reached under vacuum conditions, which dramatically restricts the application conditions of the devices.

Shortening the device channel lengths, which can increase the carrier extraction speed and reduce the carrier scattering rate, is one of the effective solutions to improve the behavior of FETs. However, on one hand, creating such a short-channel device requires highly complex fabrication techniques which are typically used in traditional semiconductor-based FETs, such as electron beam lithography (EBL), high-energy-ion implementation, and rapid high-temperature activation.¹⁸ On the other hand, because of the instability of perovskite materials to ultraviolet irradiation and a hydration environment in the above processes, they are almost unsuitable for preparing

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short-channel perovskite-based FETs. Thereby, it has remained a substantial challenge to determine whether it is possible to simultaneously achieve a high on/off ratio, high mobility, and large current density at room temperature in perovskite-based FETs.

A vertical field-effect transistor (VFET) architecture feature has emerged—with improved device capabilities, including large current density at low operating voltage and high speed, thanks to the easily reduced semiconductor channel length. The channel length is closely related to the thickness of active layer materials. In addition, different from conventional planar transistors, in which the lateral charge transport is severely limited by any cracks in the semiconductor channel, the vertical current flow in VFETs has no obvious relation with the in-plane cracks, and therefore, it can reflect the intrinsic carrier mobility of the materials.^{19,20} To date, a variety of VFETs have been investigated with a series of semiconductors, including organic semiconductors such as 2,6-diphenyl anthracene,^{21,2} fullerene,²³ as well as inorganic materials like gallium zinc oxide (IGZO),¹⁹ WS₂,²⁴ WSe₂,²⁵ MoS₂,^{26,27} PbS,^{28,29} and so on. In these device configurations, graphene, which serves as the ideal source electrode and the Fermi energy of which can be modulated by a gate electric field, can optimally modulate the performance of VFETs. And it is possible to achieve a high on/off ratio VFETs by employing graphene as a special workfunction-tunable contact for the semiconductor channel to effectively modulate the vertical current flow through the graphene-semiconductor junction.³⁰ It is important to note that with a top-electrode of Ohmic contact and a thin semiconductor channel, VFET devices can also produce a high current density suggesting possible applications in digital electronics.^{22,24,31–33} For instance, Duan and his colleagues developed a VFET by vertically stacking a P3HT/graphene contact, exhibiting good performance with a high current density of 3.4 A $\rm cm^{-2}$ and an on/off ratio over $10^{3.32}$ A graphene/BP-based VFET with a high current density >1600 A cm⁻² and a 8×10^2 on/off ratio was presented by Hersam et al.³¹ Zeng et al. reported a *p*-type perovskite-based VFET with a high current density of 12.3 A cm⁻² and on/off ratio over 10⁶; however, the device was measured under vacuum condition and the performance degraded severely at room temperature.³³ Overall, the vertical architecture opens up a fresh strategy to extract the intrinsic charge transfer property of semiconductor materials and holds enormous potential to develop high-performance optoelectronic devices.

Here, in this report, we present a strategy to construct highperformance perovskite VFETs at room temperature based on a Schottky junction formed between a monolayer graphene source contact and single-crystalline MAPbBr₃ microplate. We demonstrate that the vertical current flow through the graphene/perovskite-Schottky junction can be effectively regulated to produce a high on/off ratio over 10⁴ at ambient room temperature by employing graphene as a special, workfunction-tunable contact for the perovskite channel. Additionally, our vertical design readily allows transistors with a short channel length (determined by MAPbBr₃ microplate thickness), delivering a maximum current density of 2.69 A cm^{-2} at low working voltage, greatly exceeding those of traditional planar perovskite FETs. Furthermore, we demonstrated the dual-tunability of photoresponse of our VFET at different gate voltages and drain-source biases, respectively. Importantly, the perovskite VFETs exhibit drain-tunable rectification characteristics similar to effect of the gate modulation, which provides a

simplified strategy for use in integrated electronics and circuits based on perovskite in the future.

2. EXPERIMENTAL SECTION

2.1. Materials. The hydrobromic acid (HBr) (40% aqueous solution), methylamine (40% aqueous solution, Aladdin), lead bromide (PbBr₂, 99%), and N,N-dimethylformamide (DMF, 99.5%) were purchased from Sigma-Aldrich.

2.2. Synthesis of MABr. MABr powers were produced by the reaction of hydrobromic acid with methylamine solution. First, CH_3NH_3 and hydrobromic acid (1:1) were combined in a constant pressure funnel, swirling for 2 h in an ice–water soaking. Then the liquid was converted to white powder using rotary evaporation at 60 °C. Afterward, the product was rinsed three times each with anhydrous and absolute ethanol, respectively. Finally, the MABr powder was collected and kept at 60 °C for 1 day in a vacuum drying oven.

2.3. MAPbBr₃ Microplate Growth. The starting solution was confined between two substrates (Si/SiO₂ and glass, respectively) separated by a fixed spacer. The substrate (Si/SiO₂) and the covered glass were ultrasonically cleaned in deionized water, alcohol, acetone, and isopropanol in order to obtain cleaning surfaces. And the substrate and the covered glass were treated by a plasma cleaning process after being dried at 100 °C in a drying box. A hydrophobic surface was created by submerging the covered glass in a solution of trichloro-octadecyl silane and hexane (1:600). Next, the glass was rinsed in acetone for 30 s following by nitrogen drying. The silicon substrate was dropped with 5 μ L of the MAPbBr₃ precursor before the covering glass was placed on the top. Finally, the obtained sample was heated from 60 to 100 °C gradually.

2.4. Characterization. An Agilent Cary 5000 spectrophotometer was utilized to measure the UV–vis absorption spectrum of the MAPbBr₃ microplate. Raman spectra and PL spectra of MAPbBr₃ microplates were collected by using a HORIBA Scientific Raman spectrometer under the ambient environment. A Nikon SMZ25 stereomicroscope was utilized to capture an optical image of the microplate. A Keyence laser confocal microscope (VK-X1000) was utilized to measure the microplates' thickness. A Hitachi S-4800 was utilized to capture the cross-sectional scanning electron microscope (SEM) image. And the X-ray diffraction spectra of MAPbBr₃ was characterized with a Bruker D8 Focus.

2.5. Fabrication and Characterization of the VFET. To construct the VFET, monolayer graphene produced on the surface of copper foil by chemical vapor deposition (CVD) was first transferred to an *n*-type doped Si/SiO₂ (250 nm) substrate by a wet transfer method. MAPbBr₃ microplate was directly grown on the monolayer graphene using the above-mentioned approach. Finally, gold (80 nm) electrodes was evaporated onto the monolayer graphene as a source contact. And the drain electrode was prepared using a dry transfer technique.

With a Semiconductor Parametric Analyzer (Keithley 4200A, Tektronix) and an LED light (OptoSigma, SLA-100A) as the light source, all of the electrical and optoelectrical performance of the device was collected at room temperature under the ambient atmosphere. The power intensity of the LED light was adjustable from 10 to 70 mW cm⁻². An optical shutter (VS25S2TO, UNIBLITZ) with a 2.5 mm diameter was used to accurately control the irradiation time of the sample.

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Figure 1. Fundamental characterizations of as-fabricated MAPbBr₃ microplates. (a) Pseudocolor plot and (b) three-dimensional (3D) pseudocolor plot of the MAPbBr₃ microplate. The thickness is $3.88 \ \mu$ m. (c) The absorption and photoluminescence spectra of the MAPbBr₃ microplate. Inset: Absorption spectrum to calculate the bandgap energy. (d) Raman spectrum of the MAPbBr₃ microplate. The excitation wavelength is 785 nm. (e) Characteristic J_D -V trace of the MAPbBr₃ microplate. (f) Comparison of the mobility and trap density of the MAPbBr₃ in this work with previously reported MAPbBr₃ bulk crystals or thin films (the details as shown in the references of the Supporting Information).

3. RESULTS AND DISCUSSION

MAPbBr₃ microplates were grown from the space confined solution-processed method detailed in ref 34. In our experiments, to promote the lateral growth of the perovskite microplates, we further modified the direction of growth by restricting the initial solution between two substrates separated by a fixed spacer. Figure 1a,b shows the 3D pseudocolor plots of the MAPbBr₃ microplates with a thickness of 3.88 μ m. From these plots, we can see that the microplate's surface is smooth, and its edge is regular. The smooth surfaces without discernible grain boundaries and voids were clearly evident in the SEM image (Figure S1a, Supporting Information). And the energy-dispersive spectroscopy (EDS) examination results were persented in Figure S1b (Supporting Information), which confirmed the composition of these microplates. The EDS mapping images showed that the constituent elements were uniformly distribute. Besides, we also studied the optical absorption and photoluminescence performance of the MAPbBr₃ microplate. Figure 1c displays the absorption spectrum along with a plot of wavelength versus absorbance

(inset) used to the determine the optical bandgap. The optical absorption cutoff is shown at around 565 nm, and the calculated optical bandgap E_g is 2.20 eV for the grown perovskite. The result of microplate's X-ray diffraction (XRD) is displayed in Figure S2 in the Supporting Information. The upper facet of the MAPbBr₃ microplate shows only (100), (200), (300), and (400) diffraction peaks, indicating a highly oriented crystal structure. The Raman spectrum of the MAPbBr₃ microplate exhibits molecular vibrational peaks (60, 324, 916, 969, 1253, 1477, 1584, and 2966 cm⁻¹), corresponding to PbBr₃ inorganic cage vibrations, the MA torsional mode $[\tau(MA)]$, the MA rocking mode $[\rho_1(MA)]$, the C-N stretching mode $[\nu(C-N)]$, the MA rocking mode $[\rho_2(MA)]$, and the asymmetric/stretching of the NH₃⁺ groups $[\delta_{as}(NH_3^+)/\nu_s(NH_3^+)]$, respectively, which is in accordance with those in previous reports,³⁵ as seen in Figure 1d. Furthermore, we measured the carrier mobility and trap density of the MAPbBr₃ microplate through a space charge limiting current (SCLC) method.³⁶ The dark current J_D -V trace for the MAPbBr₃ microplate at room temperature is



Figure 2. Electrical characteristics of the perovskite-based VFET. (a) Schematic illustration of the VFET based on MAPbBr₃ microplates. (b) The output characteristics under different gate biases. (c) The band structure under (i) negative gate voltage ($V_{gs} < 0$) and (ii) positive gate voltage ($V_{gs} < 0$) at zero drain-source bias. (d) The transfer characteristics I_{ds} vs V_{gs} (black solid square) and $I_{ds}^{1/2}$ vs V_{gs} (red solid line) of the device. V_{gs} ranged from -60 to 20 V; $V_{ds} = -20$ V. (e) The statistics of carrier mobilities of 20 devices in our experiments.

shown in Figure 1e. With increasing applied voltage, in the J_D -V trace, an ohmic regime occurs at low bias (red line); a trapfilled regime (green line) starts a point (V_{TFL}) indicating all the traps are filled at this bias; and the Child regime is at high bias (orange line). The trap density (n_t) and carrier mobility (μ) could be derived by the equations below:^{36–38}

$$V_{TFL} = en_t L^2 / (2\varepsilon\varepsilon_0)$$
$$J_D = 9\varepsilon\varepsilon_0 \mu V_b^2 / (8L^3)$$

where V_{TFL} is the trap-filled limit voltage, e is the electron charge, L refers to the thickness of the MAPbBr₃ microplate (3.88 μ m), ε_0 is the vacuum permittivity, ε is relative dielectric constant of MAPbBr₃ (about 4.8),³⁹ and J_D represents the dark current density at V_b , which is the bias between trap-filled regime and Child regime. The n_t and μ for the MAPbBr₃ microplate are estimated to be 3.82×10^{10} cm³ and 100.87 cm² V^{-1} s⁻¹ respectively, which is competitive to those for the perovskite single-crystal microplates previously reported (Figure 1f). All of the above properties indicate that the MAPbBr₃ microplate has good crystallinity.

Figure 2a exhibits the relevant scheme diagram of the VFET. The bulk silicon back-gate electrode, SiO_2 gate dielectric, monolayer graphene source electrode, MAPbBr₃ microplate, and Au drain electrode are overlapped from bottom to top to construct the device. The graphene serves as bottom contact

with the active layer, and Au metal is used as the top contact. A wet transfer technique was utilized to transfer the monolayer graphene synthesized by the CVD method⁴⁰ to a prepared Si/ SiO₂ (250 nm) substrate.⁴¹ The Raman spectrum of the graphene on the Si/SiO₂ substrate is shown in Figure S3a in the Supporting Information. The graphene exhibits two most distinctive characteristics, the G peak near 1585.3 cm⁻¹, which arises from the in-plane vibration of the sp^2 hybrid carbon atom, and the 2D peak around 2701.06 cm⁻¹, which describes the interlamellar stacking of carbon atoms in graphene samples.⁴² Both peaks exhibit a perfect simple Lorentz distribution, and it is noticed that a sharp 2D peak was almost twice as high as the G peak, proving the monolayer nature of graphene with few defects.⁴³Figure S3b in the Supporting Information shows the photoluminescence (PL) spectra of the MAPbBr₃ microplate and MAPbBr₃ microplate on graphene. The good interface quality derived from the direct growth of the MAPbBr₃ microplate on monolayer graphene is favorable to the effective separation and transfer of photogenerated carriers at the perovskite/graphene interface. Both the perovskite on top of graphene and pristine perovskite exhibits a prominent PL peak at 535 nm. However, a significant PL quenching is observed at the MAPbBr₃/graphene interface, which reflects a fact that the interface coupling is enhanced in this heterostructure system. The quenching of the PL intensity indicated that the radiative emission of perovskite was



Figure 3. Dynamic regulation of device characteristics by drain-source biases and the physical mechanism. Transfer characteristics at different drain-source biases (a) in the dark state and (b) under light illumination with a power density of 13.44 mW cm⁻². (c) The energy band diagram of perovskite-based VFET with V_{ds} ranging from -2 to -20 V in the *p*-channel region. (d) The energy band diagram of perovskite-based VFET with V_{ds} ranging from +2 to +20 V in the *n*-channel region.

suppressed, which prevents the singlet excitons radiating and decaying to the ground state. $^{\rm 44}$

The electrical properties of our perovskite-based VFETs were measured at room temperature under ambient conditions. Figure 2b displays the output characteristics $(I_{ds}-V_{ds})$ of the VFET at different gate voltages, which demonstrates effective gate-modulating characteristics. It is noteworthy that the device also displays an ambipolar behavior, which was attributed to the gate-tunable Schottky barrier between the graphene and the perovskite channel layer. The working mechanism of our perovskite-based VFET can be explained by the energy band alignment (Figure 2c). As a zero-bandgap semiconductor, the work function of graphene can be tuned around the charge neutrality point. Additionally, the weak electrostatic shielding effect and low state density of graphene make the Schottky barrier height at the graphene-channel interface more sensitive to gate voltage than the Au-electrode/ perovskite channel layer interface at the top. As a result, the Schottky barrier height at the interface between graphene and perovskite can be effectively adjusted by applying gate voltage. At the negative gate voltage, the holes accumulated at the graphene/perovskite interface, and the Fermi level of graphene is lower (Figure 2c (i)). Therefore, the barrier height for hole

injection is reduced, resulting in a large negative holedominant current as promoting gate voltages. In contrast, when the gate voltage is positive, the Fermi level of graphene is elevated, creating a significant barrier that prevents the hole injection from graphene to perovskite (Figure 2c (ii)), resulting in the easier injection of electrons and change of the polarity of the drain-source current. Furthermore, the tunable Schottky barrier in the perovskite-based VFET also significantly improved the on/off ratio and current density. As a result, the perovskite-based VFET showed a U-shaped transfer curve containing both *p*-channel and *n*-channel regions at different gate voltages, V_{gs} (Figure 2d), which confirmed the ambipolar transport properties of the perovskite VFET. Figure 2d depicts the ambipolar transfer characteristics of VFET measured at room temperature with a drain voltage of -20 V as sweeping the gate voltage, along with the $I_{ds}^{1/2}$ vs V_{gs} plots. The curves are linear over a wide range of V_{gs} for both *p*-type and *n*-type channels. When the V_{gs} was swept from -60 to -27 V and the drain-source voltage (V_{ds}) is maintained at -20 V, the device operates effectively between on and off states with a large on/off ratio of 2.64×10^4 and a current density of 2.69 A cm⁻². The on/off ratio has only a slight change even at small voltages of $V_{ds} = \pm 2, \pm 10$ V (Figure S4, Supporting



Figure 4. Photoresponse characterization of the perovskite-based VFET. (a) The transfer characteristics of the device under various illumination intensities. The incident illuminant is an LED light. (b) The responsivity as a function of gate voltage with various drain-source voltages at a power density of 29.43 mW cm⁻². (c) The responsivity and detectivity as a function of drain-source voltage in *p*- and *n*-channel operating regimes with a power density of 29.43 mW cm⁻². (d) Responsivity of the photodetector as a function of incident light powers in *p*- and *n*-channel operating regimes.

Information). The carrier (electron and hole) mobilities could be determined in the saturation regime with the saturation I_{ds} vs V_{gs} relationship:

$$I_{ds} = \frac{w}{2L}C_{i}\mu(V_{gs} - V_{th})^{2}$$

where W and L stand for the channel width and length, respectively. C_i represents the gate capacitance of the dielectric layer, and V_{th} is the threshold voltage.

The statistical distribution of carrier mobility for 20 investigated perovskite-based VFETs under the consistent test conditions was summarized in Figure 2e. According to the experiment results, the maximum saturation carrier mobilities for the device are 23.39 (electron) and 14.93 cm² V⁻¹ s⁻¹ (hole), respectively. Whether in *n*-channel or *p*-channel regions, the excellent field-effect characteristics can be attributed to the following points: (i) the direction of the drain to source electrical field is parallel to the direction of the gate-induced electrical field, which can weaken the effect of ion migration;⁴⁵ (ii) the short channel of the perovskite-based VFET and the large surface area of the vertical junction can effectively facilitate the transport of electrons and holes and reduce the driving voltage, so that the device can operate with a lower voltage and power dissipation (46.11 nW).

To confirm the benefits of our VFET, we also compared traditional planar transistors with an MAPbBr₃ microplate of the same thickness, as shown in Figure S5 in the Supporting Information. The channel length of the planar transistor is 25 μ m, which is determined by the size of shadow mask during patterning and is substantially longer than that of the VFET. The output curves of the planar device were depicted in Figure S5b in the Supporting Information. The device possesses an

on/off ratio of $\sim 10^2$, nearly 2 orders of magnitude lower compared to that of the perovskite-based VFET. In the same gate voltage range, the variation of drain-source current in the planar device is obviously lower than that of the VFET. The transfer curves shown in Figure S5c,d in the Supporting Information demonstrate ambipolar transport properties as well, and the current density of the planar transistor in the *n*type and *p*-type regions is calculated to be 0.023 and 0.041 A cm⁻², respectively, which are about 2 orders of magnitude lower than that of VFET. The above results validate the optimized performance by utilizing vertical FET structure.

Interestingly, we found that our VFET device also shows tunable field-effect characteristics with varying the V_{ds} (Figure 3a). When the negative V_{ds} was increased from -2 to -20 V, the transfer curves shift toward the left; by contrast, when the V_{ds} was modulated positive, they shift to the right. That is, the transfer curve shifts horizontally in both the p-channel and nchannel regions with a larger threshold voltage as the magnitude V_{ds} increases. In order to verify these phenomena, we further measured the transfer curves under light illumination of different power densities (Figure 3b and Figure S6, Supporting Information). According to Figure 3b, the device exhibits a notable increase in output current at different V_{ds} under illumination with a power density of 13.4 mW cm⁻² in comparison to the dark-state curves. However, we found the threshold voltage almost unchanged at the same V_{ds} before and after illumination. Based on the above observations, we hypothesize that the efficient V_{ds} modulation mainly originates from the regulation of the polarity of the graphene/perovskite Schottky barrier, which can be described based on the energy band alignment as schematically shown in Figure 3c,d. Figure 3c exhibits the energy band alignment of the perovskite-based

VFET with V_{ds} ranging from -2 to -20 V in the *p*-channel region. At low V_{ds} (-2 V), the Fermi level in the graphene electrode is close to the Dirac point. Because of the lack of density of states close to the Dirac point of graphene, the gate field greatly changes the Schottky barrier of the graphene/ perovskite contact. As the V_{ds} increases, the applied drain voltage draws electrons to the graphene side in the capacitor formed by the graphene/perovskite/metal, resulting in the shift of Fermi level, E_{p} in the graphene to the deep conduction band (CB).⁴⁶ The surface of graphene is reversed from p-type to ntype, which results in the electron doping of perovskite, that is, electrons are the majority charge carrier in the device under these conditions. Therefore, the threshold voltage of the pchannel gradually increases with the V_{ds} increasing. Similarly, the regulation of V_{ds} in the *n*-channel region (in Figure 3d) is similar to that in the *p*-channel region. At low V_{ds} (+2 V), the E_f of graphene is close to the Dirac point, and as the V_{ds} increases, the holes transfer to the graphene electrode in the capacitor formed by the graphene/perovskite/metal, causing the shift of E_f in the graphene to the valence band (VB). The surface of graphene is reversed from *n*-type to *p*-type, resulting that the holes are the majority charge carrier of the device. The threshold voltage exhibits a right shift. In short, the change in V_{ds} affects the polarity of graphene, which further dopes perovskite with varying majority carrier types and transport properties.

The above enhanced performance of our perovskite-based VFETs with high on/off ratio and current density has potential applications in vertical photodetectors where signal detection and amplification can be combined in a single device. Figure 4a displays the transfer characteristics of the phototransistor under various light power densities, which shows greatly enhanced photocurrent in both *p*- and *n*-channel regions. The output current as a function of power densities at a fixed drainsource bias and a gate voltage of $V_{ds} = -2$ V and $V_{gs} = -20$ V is summarized in Figure S7 in the Supporting Information. At low power density, the photocurrent increases rapidly because the MAPbBr₃ microplate absorbs light and produces a large number of photocarriers, thus exhibiting a sharp increase in current. The photocurrent increases slowly with the further increase of power density. The approximate photocurrent saturation effect indicates the photocarriers have reached the maximum absorption in the MAPbBr₃ microplate under high illumination. The responsivity (R) and detectivity (D^*) are the key performance factors to assess the optoelectronic device. R can be calculated as

$$R = \frac{I_{light} - I_{dark}}{AE_e} = \frac{\Delta I_{ds}}{AE_e}$$

where ΔI_{ds} is the photocurrent, *A* is the effective device area, and E_e is the power density of the incident light. If assuming the dark current dominates the device noise, D^* can be determined by the following equation:

$$D^* = \frac{R}{\sqrt{2qJ_{dark}}}$$

where R refers to the responsivity, q means charge quantity, and J_{dark} corresponds to the dark current density of the photodetector. In our device, in addition to the V_{gs} , the V_{ds} can also modulate the device performance as shown in Figure 4b. The detectivity as a function of V_{gs} with various V_{ds} also indicates the efficient dual-modulation of the device photoresponse by external biases (Figure S8, Supporting Information). Figure 4c summarized R and D^* as a function of V_{ds} in pchannel and *n*-channel regions, respectively. R can be tunable from 0.28 to 4.8 A W^{1-} in the *p*-channel region and from 0.51 to 15.98 A W¹⁻ in the *n*-channel region. The maximum D^* of the device can reach up to 10^{13} Jones both in *n*-channel and *p*channel regions, due to the relatively small dark current density and the high responsivity. Simultaneously, we also evaluated the device noise spectrum and the corresponding detectivity of the device (Figures S9 and S10, Supporting Information), which exhibit slightly lower values but a similar changing trend as the results if assuming dark current dominates the device noise. It is worth noting that the modulation effect of the V_{ds} on R and D^* is different in p- and n- channel regions. The difference of R and D^* in p- and n-channel regions mainly originates from a difference in the hole and electron mobility. The mobility of electrons in the device is distinctly higher than that of holes, as shown in Figure 2e. Therefore, the R in the nchannel region is generally higher than that in the *p*-channel region under the same V_{ds} range we have measured. Under negative gate voltage (solid lines in Figure 4c), R shows an increasing trend as the V_{ds} changes from -20 to 20 V. However, as the gate voltage is positive (dashed lines in Figure 4c), R shows a decreasing trend as the V_{ds} changes from -20 to 20 V, which is attributed to the modulation of the graphene polarity by the V_{ds} . When the gate voltage is negative, the device works in the hole transport mode, and the surface of graphene is reversed from *n*-type to *p*-type as the V_{ds} gradually changes from -20 to 20 V. The hole concentration in the device changes from depletion mode to accumulation mode, resulting in the gradual decrease of the potential energy barrier between graphene and perovskite, leading to larger photocurrent. Hence, the *R* shows an increasing trend, while the dark current keeps almost consistent (observed from Figure 3b). However, when the gate voltage is positive, the device works in the electron-dominated range. As the surface of graphene reversed from *n*-type to *p*-type when the V_{ds} changes from -20to 20 V, the electron concentration in the device changes from the accumulation mode to the depletion mode; thus, the potential energy barrier between graphene and perovskite increases, and the photocurrent decreases. Therefore, the R of the device shows a decreasing trend. The method of dynamic modulation V_{ds} can achieve a wide range of device performance modulations with simplifying the device architecture (Figure 4b). Furthermore, Figure 4d depicts the relationship between *R* and power density with various V_{gs} . Under $V_{ds} = -20$ V and $V_{gs} = +20$ V, *R* and *D** were approximately 11.78 A W¹⁻ and 1.55×10^{14} Jones at a light density of 13.44 mW cm⁻² for the perovskite-based VFET in the n-channel region and then decrease as light density is promoted, which is constant with previous reports and can be ascribed to the increased recombination probability of the photogenerated electronhole pairs as the increase of power density.⁴⁷

Additionally, the photoresponse time is another crucial factor for photodetectors, which reflects the sensitivity and ensures that the device can follow the fast-switching light signal. The temporal photoresponses of the device were measured by chopper-generated light pulses at an irradiance of 13.44 mW cm⁻², as shown in Figure S7d. The device demonstrates higher dynamic properties and can be easily switched between "ON" and "OFF" states when a continuous pulse of light delivered to the device.

4. CONCLUSION

In conclusion, we have demonstrated perovskite-based VFETs with excellent performance including a high on/off ratio up to 10⁴, carrier mobility of 23.4 cm² V⁻¹ s⁻¹, and a current density of 2.69 A cm⁻² at room temperature. Additionally, we found that drain-source voltage can modulate the polarity of dominant carriers similar to that of the gate voltage in our perovskite-based VFET by regulating the source contactperovskite Schottky barrier, thus exhibiting dual-modulation properties. Meanwhile, our perovskite-based VFETs exhibited remarkable photoelectric performance with a responsivity of 11.78 A W¹⁻ and a detectivity of 10¹⁴ Jones at the light irradiance of 13.44 mW cm⁻² in the *n*-channel region. Our work develops a simple device architecture with enhanced device performance, and we believe that this high-performance perovskite-based VFET can provide a facile route for future electronic and optoelectronic applications.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsphotonics.2c01501.

SEM image, EDS color mapping, and XRD of the MAPbBr₃ microplate; Raman spectrum of the monolayer graphene; comparison of the PL spectra of the MAPbBr₃ microplate and MAPbBr₃/graphene; transfer characteristics at $V_{ds} = \pm 2$ and ± 10 V of the perovskitebased VFET; fabrication scheme and electrical characterization for the planar transistor; photoresponse and sensitivity of the perovskite-based VFET with the drainsource bias and gate voltage modulation (PDF)

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Notes

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