Heterostructures play a vital role in functional devices on the basis of the individual constituents. Non-conventional heterostructures formed by stacking 2D materials onto structurally distinct materials are of great interest in achieving novel phenomena that are both scientifically and technologically relevant. Here, a heterostructure based on a 2D (molybdenum ditelluride) MoTe₂ and an amorphous strontium titanium oxide (a-STO) thin film is reported. The heterostructure functions as a high-performance photodetector, which exhibits anomalous negative photoresponse in the pristine device due to the scattering effect from the light-induced O⁻ ions. The photoresponsivity and the specific detectivity are found to be $>10^4$ AW⁻¹ and $>10^{13}$ Jones, respectively, which are significantly higher than those in standard MoTe₂ devices. Moreover, through tuning the light programming time, the photodetection behavior of the MoTe₂/a-STO heterostructure experiences a dynamic evolution from negative to positive. This is due to the optically controllable modulation of the interfacial states, which is further confirmed by the X-ray photoelectron spectroscopy and photoluminescence measurements. It is envisioned that the 2D material/a-STO heterostructure could be a potential platform for exploring new functional devices.
Figure 1. a) Schematic illustrations of the MoTe$_2$/a-STO device and the simplified energy band structure of MoTe$_2$. The fluctuations of local potential are induced by the interfacial states in the heterostructure. b) AFM images of the as-fabricated device and the enlarged area on the a-STO thin film. Scale bar is 5 µm. c) Transport characterization of the MoTe$_2$/a-STO device under bias $V_{sd} = 100$ mV. Significant hysteresis is observed, suggesting the existence of interfacial states.

However, since the direct deposition of high-quality crystalline STO (c-STO) thin film on silicon remains challenging, most of the investigations on 2D material/STO heterostructures are restricted in the commercial bulk STO substrate, which limits the potential of integrating such heterostructure with the CMOS platform. On the other hand, amorphous STO (a-STO), which demonstrates equally extraordinary properties as its crystalline counterpart, can be conveniently grown on complementary metal–oxide–semiconductor (CMOS) compatible dielectrics (for example, SiO$_2$) using pulsed laser deposition (PLD). Therefore, 2D material/a-STO heterostructure could offer an alternative route for practical integration in heterogeneous electronic circuitry. Exploring and understanding the behavior of such heterostructure could thus open up a new avenue for designing new electronic and optoelectronic architectures.

In this work, we report a heterostructure consisting of a bilayer of molybdenum ditelluride (MoTe$_2$) and a PLD grown a-STO thin film on a Si/SiO$_2$ substrate. Structural defects on the a-STO surface cause significant modulation of the optical/electrical properties of the MoTe$_2$ through the formation of interfacial states. The MoTe$_2$/a-STO heterostructure can function as a high-performance photodetector, in which anomalous negative photodetection behavior is observed under light illumination with different wavelengths. Such photodetector exhibits remarkable enhancements in both the photoresponsivity ($>10^4$ A W$^{-1}$) and the specific detectivity ($>10^{13}$ Jones). Furthermore, we demonstrate that the interfacial states evolve in an optically controllable manner, which is represented by the light illumination induced modification in both the transport property and the photodetection behavior of the MoTe$_2$ device. Such optically tunable interfacial states evolution can also be generalized to other 2D materials, which is validated in the heterostructures of molybdenum disulfide (MoS$_2$)/a-STO, tungsten disulfide (WS$_2$)/a-STO, tungsten diselenide (WSe$_2$)/a-STO, and molybdenum diselenide (MoSe$_2$)/a-STO.

Figure 1a depicts a schematic of the MoTe$_2$/a-STO heterostructure fabricated in a two-terminal field-effect-transistor configuration. An a-STO thin film with a thickness of ≈15 nm is directly deposited on a commercial SiO$_2$/Si substrate using a standard PLD method (see the Experimental Section for details). In contrast to the bulk crystalline STO, the a-STO film exhibits a corrugated surface morphology with a plateau-like feature as shown in Figure 1b, indicating the existence of significant amounts of structural defects. Such structural defects are considered as an effective tailoring factor at the MoTe$_2$/a-STO interface, introducing interfacial states that locally modulate the energy band structure of MoTe$_2$, which is represented by the local potential fluctuations (Figure 1a). We also conduct contact angle measurement to qualitatively visualize the interfacial states. As shown in Figure S1 (Supporting Information), the contact angle of the a-STO surface is 46.5°, which is effectively smaller than that of the SiO$_2$ substrate (63°), suggesting a more hydrophilic surface property caused by the tailoring states. The significant modification of the interfacial properties could induce considerable variation.
in both the electrical and optoelectrical behavior of the MoTe₂ device.\[15–17]\] The optical image of the as-fabricated device is shown in Figure S2a (Supporting Information), with the 2D MoTe₂ crystal confirmed by Raman spectroscopy (Figure S2c, Supporting Information). The thickness of the MoTe₂ flake is revealed by the atomic force microscopy (AFM) line profile as \( \approx 1.54 \) nm, indicating its bilayer nature (Figure 1b). The corrugations induced by the a-STO film can also be viewed in the 3D AFM image (Figure S2b, Supporting Information). Note that considerable surface corrugations are observed even in the MoTe₂ covered area, indicating that the MoTe₂ flake effectively follows the a-STO surface morphology without semisuspension on the spikes. The basic transport characterization of the heterostructure is demonstrated in Figure 1c, displaying a typical n-type dominated behavior. An enhanced hysteresis loop is observed in the transfer curve as compared with the MoTe₂ device fabricated on standard SiO₂ substrate (Figure S3a, Supporting Information), which further confirms the effect of the interfacial states. Nevertheless, as shown in Figure S4a (Supporting Information), the output characterization at multiple gate voltages still exhibits nearly ideal linear relationship, illustrating the negligible influence of the interfacial states on the quality of the electrical contacts.

Since the photodetection behavior of the 2D materials has been proved to be sensitive to the interfacial states,\[18,19]\] we utilize the MoTe₂/a-STO heterostructure as a photodetector to further explore its properties. We first characterize the bare a-STO substrate to exclude the possibility of its intrinsic photoresponse (Figure S5, Supporting Information). Figure 2a shows the output curves of the heterostructure photodetector under both dark and illumination (\( \lambda = 638 \) nm) conditions at a backgate voltage of \( V_g = 0 \) V. A lower channel current under illumination is observed through the entire window of applied source–drain bias \( V_{sd} \), indicating a negative photodetection behavior. The photocurrent \( I_{ph} \) at each \( V_{sd} \) is extracted using \( I_{ph} = I_{light} - I_{dark} \), where \( I_{light} \) and \( I_{dark} \) are the source–drain currents under light illumination and dark condition, respectively, and a linear dependence on the \( V_{sd} \) is demonstrated (Figure S4b, Supporting Information). To further illustrate the photodetection property of the MoTe₂/a-STO photodetector, we investigate the \( I_{ph} \) at various \( V_g \) and light illumination power (Figure 2b,c). The negative \( I_{ph} \) is consistently observed at each \( V_g \) under different illumination power, suggesting a universal photodetection characteristic in the pristine MoTe₂/a-STO heterostructure. Note that the saturated \( I_{ph} \) at higher \( V_g \) is due to the saturation of the transport current as shown in the transfer characterization (Figure 1c). The stability of such a negative photoresponse is also investigated, as shown in Figure S6 (Supporting Information). We note that previous works, in general, reveal a positive \( I_{ph} \) in the MoTe₂ phototransistors fabricated on SiO₂ substrate, which originates from the introduction of photocarriers in MoTe₂ under illumination.\[20,21]\] This is distinct from the photoresponse behavior in our MoTe₂/a-
STO heterostructure transistor, in which negative photocurrent is identified at all the biases and gates. Such negative photoresponse behavior is also in contrast to that in a system dominated by the interfacial states, where positive photodetection with relatively long response time is expected due to the charge trapping induced photogating effect.\(^{[22,23]}\)

Moreover, the photocurrent generation in the MoTe\(_2\)/a-STO device is significantly improved compared with the SiO\(_2\) supported MoTe\(_2\) photodetector measured in the same conditions. To quantitatively illustrate the performance of the MoTe\(_2\)/a-STO photodetector, we determine the photoresponsivity as 
\[
R = \frac{I_{ph}}{P \cdot S},
\]
where \(P\) is the light intensity, and \(S\) is the effective area under illumination. As shown in Figure 2d, the photoresponsivity of the MoTe\(_2\)/a-STO photodetector reaches up to \(10^4\) A W\(^{-1}\) as decreasing light power density, demonstrating more than two orders of magnitude enhancement on the basis of a standard MoTe\(_2\)/SiO\(_2\) device (Figure S3d, Supporting Information). The current noise spectra at multiple \(V_g\) are depicted in Figure 2e, the noise equivalent power (NEP) is then calculated as 
\[
\text{NEP} = \text{noise density} / R.
\]

The detailed information about how to measure the current noise spectra of the MoTe\(_2\)/a-STO heterostructure is demonstrated in Figure S7 (Supporting Information). The NEP saturates at \(\approx 2 \times 10^{-5}\) pW Hz\(^{-1/2}\) with increasing \(V_g\), suggesting the great potential of applying MoTe\(_2\)/a-STO heterostructure in ultra-low light power detection (Figure S8, Supporting Information). We also investigate the gate-dependent behavior of the specific detectivity defined as 
\[
D* = \frac{(B \cdot S)^{1/2}}{\text{NEP}},
\]
where \(B\) is the measuring bandwidth. The value of \(D*\) peaked at \(V_g = -10\) V, followed by a gradual saturation at \(\approx 10^{13}\) Jones (Figure 2f), which is considerably higher than those reported in other 2D materials.\(^{[24–26]}\) Additionally, the photodetection behavior of the MoTe\(_2\)/a-STO heterostructure at other wavelengths (\(\lambda = 515, 473,\) and \(405\) nm) is demonstrated in Figure S9 (Supporting Information), in which negative photoresponse and remarkably enhanced photodetection performance are consistently observed. The wider spectrum photoresponse of the heterostructure is also investigated (Figure S10, Supporting Information), where the \(R\) gradually decreases with increasing wavelength. Nevertheless, the \(R\) can still be maintained in the magnitude of \(\approx 10^{3}\) A W\(^{-1}\), indicating the potential of using such heterostructure for broadband photodetector.

To obtain comprehensive information on the photoresponse behavior of the MoTe\(_2\)/a-STO heterostructure, we dynamically monitor the device performance under several consecutive light illumination periods with the duration fixed at 75 s, and the gate voltage \(V_g\) is set to be negative within each illumination process. Figure 3a shows the time-dependent photocurrent of the MoTe\(_2\)/a-STO device. Thenegative photocurrent with decreasing magnitude is persistently observed until the total illumination time \(t_{pro} = 450\) s. However, when the \(t_{pro}\) reaches 600 s, the polarity of the photocurrent switches to positive. Moreover, the evolution of the photocurrent is accompanied by the shortening of the photoresponse time, which finally approaches the intrinsic fast photoresponse speed of a standard MoTe\(_2\) photodetector. In addition to the photoresponse behavior, the transport characterization after each illumination period was also investigated (Figure 3b), which exhibits a significant electron doping effect with the weakened hysteresis as increasing the illumination time. The evolution of the transport characterization suggests a photogating effect that has been previously observed in the 2D photodetectors with the assistance of interfacial states.\(^{[27,28]}\) During the illumination process, the photogenerated holes are continu-
Figure 4. a) Schematic illustration of the evolution of the interfacial state of the MoTe₂/a-STO device. The green spheres indicate the traps at the interface, while the pink ones are the charged oxygen atoms. The red and blue circles represent the photogenerated hole and electron, respectively. In the pristine device, the light-induced O\(^{\delta^-}\) scattering centers lead to the initial negative photocurrent. After light programming under \(V_g < 0\), the photogenerated holes in MoTe₂ are continuously trapped at the interfacial states, which screens the scattering effect arising from the O\(^{\delta^-}\) ions. In the intermediate stage 1, the negative photocurrent caused by scattering is balanced with the intrinsic positive photocurrent, therefore, no photocurrent is detected in the device. A positive photocurrent is finally observed when the O\(^{\delta^-}\) ions are completely screened by the trapped holes, as shown in stage 2. b) XPS spectra of the Sr 3d, Ti 2p, and O 1s core levels in the a-STO thin film. The emergence of Ti^{3+} peaks indicates the existence of the oxygen vacancies, which is further confirmed by the O 1s core level. c) PL spectrum of the a-STO thin film. The peak located at 612 nm demonstrates the oxygen vacancy induced gap states, which is consistent with the XPS spectra.

Initially, the O\(^{\delta^-}\) ions created by the light illumination are able to migrate within the a-STO film or move toward the MoTe₂/a-STO interface. These O\(^{\delta^-}\) ions behave as the scattering centers to degrade the MoTe₂ channel current, which is represented by the negative photocurrent. When the light is off, the photogenerated electrons recombine with the charged gap states, and the O\(^{\delta^-}\) ions revert to oxygen atoms that are reabsorbed into the a-STO film, the channel current is thus restored due to the

Additionally, the light-controlled evolution of the photoreponse behavior under other gate voltages are investigated, where the photocurrent possesses a consistent polarity switching as increasing the illumination time (Figure S12a, Supporting Information). Such polarity switching is also observed at other wavelengths (Figure S13a, Supporting Information). Note that the photoreponse time depends only on the illumination process, with little effects from both the gate voltage and the wavelength (Figures S12b and S13b, Supporting Information). We also investigated the a-STO with different thicknesses from 15 to 50 nm, incorporating with MoTe₂ from 1.5 nm to around 10 nm (Figures S14 and S15, Supporting Information). The electrical transport and evolution of photoreponse are consistent for the a-STO substrates and the MoTe₂ samples with various thicknesses. These results indicate that the evolution behavior of the MoTe₂/a-STO heterostructure is largely determined by the conditions of the interfacial states, which can be effectively modulated by controlling the light illumination procedure.

Although the photodetection evolution in the MoTe₂/a-STO heterostructure can be described by the light-induced changes of the interfacial states, understanding its initial anomalous negative photoreponse is essential to explore the real application of such heterostructure. It has been demonstrated in the previous work that the as-deposited STO thin film could release oxygen atoms under light illumination.\(^{[29,30]}\) Moreover, the oxygen vacancies in PLD-grown STO film could introduce optically-active gap states that provide photogenerated electrons, particularly in the case of amorphous STO thin film.\(^{[31,32]}\) These released oxygen atoms can then trap the photoexcited electrons to form negatively charged oxygen ions O\(^{\delta^-}\), where \(\delta\) is the effective charge on each oxygen atom.\(^{[30]}\) We, therefore, propose a model to synergistically describe the initial and evolved photodetection behavior in the MoTe₂/a-STO heterostructure, as shown in Figure 4a. Initially, the O\(^{\delta^-}\) ions created by the light illumination are able to migrate within the a-STO film or move toward the MoTe₂/a-STO interface. These O\(^{\delta^-}\) ions behave as the scattering centers to degrade the MoTe₂ channel current, which is represented by the negative photocurrent. When the light is off, the photogenerated electrons recombine with the charged gap states, and the O\(^{\delta^-}\) ions revert to oxygen atoms that are reabsorbed into the a-STO film, the channel current is thus restored due to the
disappearance of the scattering centers. Second, during the light illumination process, while applying negative $V_g$, the photogenerated holes in MoTe$_2$ are continuously trapped at the interfacial states under the external electric field, leading to the electron accumulation and strong n-type doping effect in the MoTe$_2$ channel. The band alignment diagram is shown in Figure S16 (Supporting Information) to explain the hole trapping and transporting in the light programming process. These trapped holes can also screen the scattering effect arising from the O$^{2-}$ ions, thus accelerating the charge separation/recombination, which results in the reduction of negative photocurrent and photoresponse time while increasing the light programming time. Similarly, in the final stage after a sufficiently longtime illumination, significant amounts of holes are trapped at the interface and completely screen the O$^{2-}$ scattering centers. As a consequence, the photodetection behavior of the MoTe$_2$/a-STO heterostructure approaches that of a heavily n-doped standard MoTe$_2$ device, where positive photocurrent with a fast response speed is expected. It is of note that the positive photocurrent will gradually decrease, and eventually evolves to the negative polarity when the retention time is 8 h (Figure S17, Supporting Information), suggesting the gradual release of the trapped holes. Upon prolonging the time, the amount of trapped holes progressively decreases, which is not sufficient to screen the scattering effect from the O$^{2-}$ ions, leading to the change of the polarity from positive to negative. Additionally, applying an opposite gate could accelerate the release of the trapped holes, which reverses the photocurrent polarity in a shorter time (Figure S18, Supporting Information).

To validate the existence of oxygen vacancies in the a-STO film, we perform the X-ray photoelectron spectroscopy (XPS) measurement on the as-deposited a-STO thin film, with all the spectra fitted with Gaussian/Lorentzian mixed functions (Figure 4b). The locations of the two Sr 3d core level peaks are consistent with previous reports, indicating that the oxidation state of Sr is insensitive to the phase of the STO film.$^{[33,34]}$ However, the Ti 2p spectrum demonstrates two additional peaks on the basis of the Ti$^{4+}$ oxidation state, suggesting the appearance of the Ti$^{3+}$ interfacial state due to the emergence of the oxygen vacancies.$^{[35,36]}$ The corresponding O 1s spectrum also reveals an extra peak located at the binding energy, further confirming the significant amounts of oxygen vacancies in the a-STO film.$^{[37]}$ On the other hand, to illustrate the oxygen vacancy induced gap states, we investigate the photoluminescence (PL) spectrum of the a-STO film at room temperature (Figure 4c). A PL peak centered at wavelength $\lambda = 612$ nm is observed, which is absent in a commercial STO substrate, indicating the existence of gap states. Note that the PL peak position is consistent with the red illumination band caused by oxygen vacancies in STO film.$^{[38]}$ These results validate our model for describing the photodetection behavior of the MoTe$_2$/a-STO heterostructure.

2D materials beyond MoTe$_2$ share a similar layered structure with varied electronic and optoelectronic properties. To substantially explore the potential of 2D material/a-STO heterostructure, we investigate MoS$_2$/a-STO, WS$_2$/a-STO, WSe$_2$/a-STO, and MoSe$_2$/a-STO heterostructures, respectively. The optical image of the as-fabricated MoS$_2$/a-STO heterostructure transistor is shown in Figure S19a (Supporting Information). The Raman spectrum of the MoS$_2$ flake reveals two characterization peaks with a separation of 21.5 cm$^{-1}$, indicating the bilayer nature of the flake (Figure S19b, Supporting Information). We observe similar photodetection behavior in the pristine MoS$_2$/a-STO device as compared with that of the MoTe$_2$/a-STO heterostructure, as illustrated in Figure 5a,b. Moreover, the transport and photoresponse properties of the MoS$_2$/a-STO device demonstrate consistent evolution characteristics that are controlled by the light illumination process, indicating the existence of the progressively evolved interfacial states (Figure 5c,d). Additionally, the WS$_2$/a-STO, WSe$_2$/a-STO, and MoSe$_2$/a-STO heterostructures also exhibit photoresponse behavior that is similar to both the MoTe$_2$ and MoS$_2$ (Figures S20–S22, Supporting Information), implying that the optically controlled evolution of interfacial states is generally applicable in 2D material/a-STO heterostructures. On the other hand, the photoresponsivity of these three photodetectors are determined as $2.9 \times 10^7$ A W$^{-1}$ (MoS$_2$), $4.3 \times 10^7$ A W$^{-1}$ (WS$_2$), $5.9 \times 10^7$ A W$^{-1}$ (WSe$_2$), and $3.1 \times 10^7$ A W$^{-1}$ (MoSe$_2$), respectively, promising the 2D material/a-STO heterostructure as a potential platform toward high-performance photodetector applications.

In summary, we have fabricated a heterostructure based on the 2D MoTe$_2$ flake and PLD grown a-STO thin film that functions as a high-performance photodetector and demonstrated optically controlled photodetection behavior driven by the evolution of interfacial states. The pristine device reveals anomalous negative photocurrent, where the photoresponsivity and specific detectivity reach up to $\approx 4 \times 10^4$ A W$^{-1}$ and $\approx 10^{13}$ Jones, respectively, which are remarkably higher than those of the MoTe$_2$ device on standard SiO$_2$ substrate. Furthermore, we demonstrate that the photoresponse behavior of the MoTe$_2$/a-STO heterostructure is highly tunable with light programming time, where the photocurrent polarity can be effectively modulated from negative to positive with two orders of magnitude to shorten the photoresponse time. The initial negative photocurrent originates from the scattering effect caused by the light-induced O$^{2-}$ ions in the a-STO film, while the light controllable photosresponse evolution is realized by the continuously trapping of photogenerated holes from MoTe$_2$ at the interfacial states. Such light-tunable photodetection behavior can also be generalized to other 2D materials, which is confirmed in the heterostructures of MoS$_2$/a-STO, WS$_2$/a-STO, WSe$_2$/a-STO, and MoSe$_2$/a-STO. Our work promises 2D material/a-STO architecture as a potential candidate for the integration into CMOS compatible platform, envisioning their opportunities for designing new heterogeneous integrated electronics.

**Experimental Section**

**PLD Growth of a-STO Thin Film on SiO$_2$/Si Substrate:** The a-STO thin film was deposited on SiO$_2$/Si substrate by pulsed laser deposition using a KrF excimer laser ($\lambda = 248$ nm) with a repetition rate of 2 Hz and laser fluence of 1.3 J cm$^{-2}$. The growth temperature and oxygen partial pressure were 640 °C and 10 mTorr, respectively. After deposition, the sample was cooled down to room temperature with a cooling rate of 10 °C min$^{-1}$ in the deposition pressure. Commercial STO single crystal was used as target. The growth rate was calibrated with the in situ reflection high-energy electron diffraction oscillations obtained during the growth of single-crystalline STO thin film on TiO$_2$-terminated STO (001) substrate.

**Device Fabrication and Characterization:** The MoTe$_2$, MoS$_2$, WS$_2$, WSe$_2$, and MoSe$_2$ flakes were mechanically exfoliated from their bulk crystals (HQ graphene) using a Scotch tape and transferred onto the PLD
grown a-STO substrate. The exfoliated flakes were located by using a high-resolution microscope (Nikon Eclipse LV100D) followed by the spin coat of polymethyl methacrylate (PMMA) photoresist. The transistor channel was defined using the conventional e-beam lithography technique (EBL). After lithography, metal electrodes Ti (5 nm) and Au (80 nm) were thermally deposited on the flakes in a high vacuum chamber ($\approx 10^{-7}$ mbar). The devices were then lifted off in acetone solution followed by the wire-bond onto a LCC chip carrier. The chip carrier was then loaded in a custom designed high vacuum system ($\approx 10^{-8}$ mbar) for electrical and optoelectrical measurements. The electrical and optoelectrical measurements were conducted using an Agilent 2912A source measure unit. Four laser beams (638, 515, 473, and 405 nm) and an exon light source configured with a monochromator were used to illuminate the device. The light density was calibrated by THORLABS GmbH (PM 100A) power meter. To acquire the noise spectra, the source terminal of the as-fabricated transistor was dc-coupled to Stanford Research SR570 low noise current preamplifier. The output of this current amplifier was recorded by an HP 35670A dynamic signal analyzer. The frequency ranges from 1 to 100 Hz for each noise spectrum measurement.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Keywords
amorphous strontium titanium oxide (a-STO), MoTe$_2$/a-STO heterostructures, optically controllable interfacial states, polarity tunable photore sponses

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