Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky Self-Powered Solar-Blind Photodetector With Robust Responsivity

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Abstract—As a 2D material, MXene has emerged as an excellent electrode material for optoelectronic devices due to its high conductivity and hydrophilic surface. Here, the Ti$_3$C$_2$-based MXene was employed to construct the Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky junction photodetector. The fabricated device demonstrated a self-powered operation manner with an extremely low dark current (0.07 pA), an outstanding light on/off switch ratio (2.5×10$^8$), a remarkable photo-response speed (43 ms/145 ms), a responsivity (R) of 15.5 mA/W, an external quantum efficiency (EQE) of 7.5% and a detectivity (D$*$) of 2.15×10$^{11}$ Jones. Such excellent photodetection performance that is comparable or even higher than those of Ga$_2$O$_3$ Schottky photodetectors previously reported is originated from the excellent conductivity of MXene, good crystallization of ε-Ga$_2$O$_3$, and their well-matched energy level. Additionally, our Schottky junction device is capable of sensing solar-blind UV region and exhibits excellent stability in the air environment. The perfect combination of 2D MXene and wide-bandgap ε-Ga$_2$O$_3$ proposes a novel route for the self-powered Schottky devices.

Index Terms—MXene, ε-Ga$_2$O$_3$, self-powered photodetector, Schottky junction.

I. INTRODUCTION

THE emergence of 2D materials and their heterostructures with fascinating electronic and optical properties provided an additional opportunity for building optoelectronic and electronic devices [1], [2]. A new branch of 2D materials, including transition metal carbides, carbonitrides, and nitrides, namely MXenes, was initially discovered by Barsoum et al. in 2011 [3]. The excellent performances such as high conductivity and hydrophilic surface indicate that MXenes can be good candidates for electronics and optoelectronics materials [4], [5]. The 2D MXenes based 2D-2D and 2D-3D van der Waals (vdW) heterostructures, are of unique ability to interact with other materials without being restricted by lattice matching, hence have recently attracted intense interest. However, the further applications of 2D-2D vdW heterostructures in large-scale and high-integrated devices is severely suffered from the small size and the complicated transfer process [6]. Compared to the 2D–2D vdW heterostructures, the 2D–3D vdW heterostructures that combine the advantages of 2D materials with the desired functions of the developed semiconductors are ideal in promising applications. In particular, MXenes based 2D-3D vdW heterostructures such as MXene/silicon [7], MXene/perovskite [8], and MXene/GaAs [9], etc. have been marginally designed for photodetectors (PDs) [10]. Inspired by all of these, reasonable selection of the semiconductors and serious-minded design of the interface are of the great significance toward exploring excellent 2D heterostructures for advanced performance PDs [11].

As an ultra-wide bandgap semiconductor, Ga$_2$O$_3$ sparks wide interest for the building of solar-blind PDs because it holds several advantages over other materials, such as instinct solar-blind UV absorption with a wide bandgap (≈4.9 eV), excellent thermal and chemical stability [12]–[20]. Ga$_2$O$_3$ has five phases of α, β, γ, δ and ε. Among these polymorphs, the β-phase is investigated most extensively for optoelectronics. In contrast, other metastable phases have been studied less, but they also have unique application characteristics in device applications [21]. The ε phase Ga$_2$O$_3$ has hexagonal and orthorhombic crystal structure. Therefore, the ε-Ga$_2$O$_3$, as a kind of Ga$_2$O$_3$ isomer, is increasing of great interest [22]. For instance, Chao and Mishra revealed that a two-dimensional electron gas is formed at the ε-Ga$_2$O$_3$/CaCO$_3$ interface with a charge density of 10$^{14}$ cm$^{-2}$, which is two orders of magnitude higher than...
that formed at the (Al$_x$Ga$_{1-x}$)$_2$O$_3$/$\beta$-Ga$_2$O$_3$ interface [23]. In addition, $\varepsilon$-Ga$_2$O$_3$ UV PDs have been investigated by lots of research groups, including ours, and these PDs have significant UV detection capabilities [24]–[27].

Here, based on the MXene/$\varepsilon$-Ga$_2$O$_3$ Schottky junction heterostructure, a self-powered solar-blind UV PD with high responsivity (15.5 mA W$^{-1}$), ultralow dark current (0.07 pA), and high light on/off ratio (2.5 $\times$ 10$^6$) under zero biasing voltage at 254 nm was devised. Moreover, to get more understanding into the inherent mechanism of the PD, the dependence of response time on the light intensity and running modes has been investigated in detail. Also, the device without packaging already exhibits excellent stability after exposure under ambient temperature for 45 days, showing the great potential of using MXene/$\varepsilon$-Ga$_2$O$_3$ Schottky junction as a promising low energy-cost self-powered solar-blind UV detectors or solar-blind UV photocells.

II. EXPERIMENTS

A. $\varepsilon$-Ga$_2$O$_3$ Film Growth

An $\varepsilon$-Ga$_2$O$_3$ layer was epitaxially grown on a (0001) sapphire substrate, carrying via a customized MOCVD growth system with a reaction chamber. Triethylgallium (TEGa) and O$_2$ were taken as gallium and oxygen precursor, respectively. TEGa and Oxygen gas were transported by the carrier gas (Ar) into the growth chamber. The growth process was carried out under the conditions of 25 Torr pressure and 470 °C temperature, and maintain 60 mins.

B. Synthesis of Ti$_3$C$_2$

Ti$_3$AlC$_2$ (MAX phase) powder (1 g) was dispersed in a mixture solution of 9 M HCl (20 mL) and LiF (1 g), followed by stirring at room temperature for 24 h to etch Al atom. The acidic suspension was washed with deionized water via centrifugation (3500 rpm for 3 min) several times, until pH ≥ 6. The resulting suspension was redispersed in deionized water and then centrifuged at 3500 rpm for 1 h. The suspension containing single-layer 2D Ti$_3$C$_2$ nanoflakes was obtained and further used for later drop-casting.

C. Device Preparation

After the epitaxial deposition of the $\varepsilon$-Ga$_2$O$_3$ film, the colloidal solution of prepared Ti$_3$C$_2$ was drop casted on the $\varepsilon$-Ga$_2$O$_3$ surface. After drying at 40 °C for 10 h, the silver and indium electrodes were fabricated on Ti$_3$C$_2$ and $\varepsilon$-Ga$_2$O$_3$ thin films, respectively (Fig. 2a). The distance between the In electrode and Ti$_3$C$_2$ is 308 $\mu$m.

D. Characterization and Measurement

The crystal structure and micromorphology features of the as-prepared samples were obtained using X-ray diffraction (XRD, D8 Advance, Bruker), scanning electron microscopy (SEM, S4800, Hitachi) and transmission electron microscopy (TEM, Tecnai G2 F30, FEI), respectively. The optical absorption spectrum measurement of the film was tested by UV--vis spectroscopy (UV-1800PC, China). The spectroscopic photoresponse of the device was measured using a monochromator combined. The photoelectric performance was recorded using a UV lamp as a light source, and a semiconductor characterization system (Keithley 4200), respectively.

III. RESULTS AND DISCUSSION

The XRD pattern shows diffraction peaks at 19.11°, 38.83° and 59.83° (Fig. 1a), which are assigned to the (002), (004) and (006) planes of the orthorhombic crystal structure of the $\varepsilon$-Ga$_2$O$_3$ film, respectively. The value of FWHM for (002) plane is 0.145. The result indicates a (002)-single preferred orientation...
of the ε-Ga$_2$O$_3$ film. In addition, the FWHM of ε-Ga$_2$O$_3$ (002) plan is 2242 arcsec obtained from the rocking curve (Fig. S1). The SEM image is also acquired on the surface of the sample as shown in Fig. 1b, indicating a fine-grained morphology of the thin film. d. The thickness of ε-Ga$_2$O$_3$ film is 350 nm measured from the cross-section SEM, as shown in Fig. S2.

The UV-vis absorption spectrum of the prepared ε-Ga$_2$O$_3$ film is shown in Fig. S3, from which the obtained band gap ($E_g$) of the ε-Ga$_2$O$_3$ film is 4.92 eV by extrapolating the line of the Tauc diagram to the x-axis of photon energy (inset of Fig. S3).

For investigations of phase information of Ti$_3$C$_2$ MXene sheets was spread casted on Si substrate. The corresponding XRD spectra of Ti$_3$C$_2$ MXene sheets are shown in Fig. 1c, where the peaks of (002) and (004) planes are found at 2θ = 6.9° and 14.0°, respectively. Besides, the morphology of the Ti$_3$C$_2$ sheets was characterized by using TEM as shown in Fig. 1d.

The schematic diagram of the Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky junction PD is presented in Fig. 2a. The inset of Fig. 2a shows the schematic of Ti$_3$C$_2$, where the purple and gray balls correspond to Ti and C atoms, respectively.

Fig. 2b depicts the I–V behavior of the Schottky PD, the dark current of the Schottky PD is ultralow (∼0.07 pA at -5 V), which is significantly lower compared to those of the Schottky junction Ga$_2$O$_3$-based PDs in previous reports[51]. First, such a low dark current can be attributed to a relatively low concentration of defects in the high crystalline quality ε-Ga$_2$O$_3$ film. Second, the formation of a “space charge region” (SCR) between the surface of Ti$_3$C$_2$ MXene and ε-Ga$_2$O$_3$ might lead to a surface barrier, which could also reduce the dark current. The value of the photo-dark current ratio ($I_{ph}/I_D$) is $2.5 \times 10^6$, confirming that the device has a high signal-to-noise ratio and high-quality photosensitive switching characteristic.

The working mechanism of the Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky PD could be explained from the energy band configuration, as illustrated in Fig. 2c, where $\Phi_b$ is the Schottky barrier height, $E_{F1M}$ is the Fermi-level of the Ti$_3$C$_2$ MXene, $E_{F2S}$ is the Fermi-level of the ε-Ga$_2$O$_3$, $E_C$ is the conduction band, $E_V$ is the valence band, and $\Phi_{bG}$ is the built-in potential. The work function ($W_M$) of the Ti$_3$C$_2$ is 4.6 eV and the electron affinity ($\chi$) of Ga$_2$O$_3$ is 4 eV, respectively, so the value of $\Phi_b$ is 0.6 eV [28], [29]. The $E_{Ga2O3}$ and Ti$_3$C$_2$ generate a Schottky junction and form an SCR in the ε-Ga$_2$O$_3$ side. The Schottky junction is dominated by a majority of carrier transport. The direction of the built-in electric field is pointing from ε-Ga$_2$O$_3$ to Ti$_3$C$_2$. Under light illumination, the photon-generated carriers in or near the SCR are separated and conducted by the internal driving force (built-in electric field).

In this configuration, the holes are transported to Ti$_3$C$_2$, while the electrons are collected by the In electrode through ε-Ga$_2$O$_3$, forming the photocurrent.

Another important characteristic of the PD is the dependence of photocurrent with light intensity. The current-voltage characteristics of the device at 254 nm illumination of different intensities are shown in Figs. S4 and 3a (high resolution). The results indicate that the photocurrent of the Schottky junction PD has a strong dependence on the light intensities. The photocurrent increases with the illumination intensity, since the density of the photoexcited carrier is proportional to the luminous flux absorbed.

Fig. 3b shows the photosresponse switching behavior of the Schottky junction PD under repeated light on/off cycles at 0 V under illumination with intensities varying from 0.1 µW/cm$^2$ to 2000 µW/cm$^2$. Even under the ultralow light intensity of 0.1 µW/cm$^2$, the photo-to-dark current switch ratio of the photo signal is 5, indicating an excellent photosresponse capability under the weak signals.

Fig. S5 shows the incident optical power-dependent output electrical signal. The dependence is given by $I_{ph} = kP^\theta$, where, $I_{ph}$ is the photocurrent, $k$ is constant for a particular wavelength, $P$ is the input optical power, the $\theta$ is the empirical value related to the complex process of trapping, and recombination within the device, which was estimated as 0.818 by fitting. This linear variation of the photocurrent suggests that the Schottky junction PD has obvious intensity-dependent features and can be fabricated for accurate solar-blind UV detection.

The response time is the capacity of a PD to respond to a changing incident light signal. Generally, it is defined as “ON” and “OFF” or “recovery” times. The devices operated in photovoltaic and photoconductive modes are also expected to have different transient light switching characteristics. We compared the performance of photo-switching characteristics of the Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky photovoltaic PD with metal-semiconductor-metal (MSM) pristine In/ε-Ga$_2$O$_3$/In conductive PD (Fig. 4a). The distance of In electrodes of conductive device is about 280 µm. Fig. 4b shows the transient photocurrent response at 254 nm for the two types of PDs with a bias voltage of 0 V and -0.1 V, respectively. In order to accurately compare the response speed of the two devices at the same photocurrent (10 nA), the Schottky device and the conductive device were tested under 400 µW/cm$^2$ and 600 µW/cm$^2$ UV illumination, respectively. In the absence of light irradiation, the Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky PD shows an ultralow dark current, only at the pA level. Even without an external bias voltage, when the light is “turned on”, the photocurrent reached ∼nA; while the signal is “turned off”, the current rapidly returns to the initial value. The results clearly show that Schottky PD has obvious and stable transient photo-response characteristics. The rise/decay times are obtained by fitting the curves using the formula (1a) and
where $I_0$ is the value of current when illumination is in turned on/turned off states, and $\tau_r$ and $\tau_d$ are the rise time and decay time. The values of the rise and decay times are fitted to be 215 and 341 ms for Schottky PD (Fig. 4c), 431 and 439 ms for conductive PD (Fig. 4d), respectively. Compared with the conductive PD without Ti$_3$C$_2$ electrode, the response speed of the Schottky PD with Ti$_3$C$_2$ nanosheets electrode is improved and faster, which shows that using Ti$_3$C$_2$ as Schottky contact causes an obvious improvement of the response speed of the device because of the construction of the built-in potential.

In addition to operating under the photovoltaic mode, Schottky PD could work in the photoconductive mode, i.e., under applied voltage. To further analyses the response times of the Schottky PD under both the forward voltage and reverse voltage, the transient response of the Schottky photovoltaic PD under different illumination power densities with a bias voltage of 0 V and -0.1 V, respectively. The response speed has a similar tendency with power density, in which the $\tau_r$ and $\tau_d$ decrease with the illumination intensities, a phenomenon attributed to the trap filling. The photocurrent transients are influenced by the filling of traps, which are-most pronounced when the injection level is raised with increasing photon flux. The most remarkable effect of the higher light density is the shortening of the rise and decay time. This general observation is due to the filling of deep trap states which lose their influence in the early stage of the photocurrent transient. This means that trap filling is more easily excited at stronger optical intensities, and hence improves the transport behavior (response speed) [32], [33]. Apparently, as the optical powers increase, the response time for Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky PD decreases faster than that of In/ε-Ga$_2$O$_3$/In conductive PD. The built-in electric field at the Ti$_3$C$_2$/ε-Ga$_2$O$_3$ interface is essential to improve separation and transportation of the photogenerated electrons and holes, and therefore promoting a more rapid response speed. The results were consistent with those shown in Fig. 4(b, c, d).

Responsivity (R), external quantum efficiency (EQE), and special detectivity (D*) are critical FOMs (Figure of Merit) for PDs and are related to each other, which is provided in Fig. 6c.

The responsivity of PD can be written as

$$ R = \frac{I_p - I_d}{P \times S} $$

Here $I_p$, $I_d$, $P$, and $S$ represent the photocurrent, dark current, light intensity, light signal exposed area, Fig. 6c shows the $R$ vs light intensities plot, which decreases with increasing light intensities. The high responsivity (15.5 mA/W) at lower illumination intensity (0.1 μW/cm$^2$) indicates outstanding performance in the sensing of low optical power. The responsivity of our PD is higher than other Schottky junction Ga$_2$O$_3$ PDs [8]–[12], because of the high crystallization of ε-Ga$_2$O$_3$ and excellent conductive of Ti$_3$C$_2$.

The EQE, which is used to describe the ratio number of electrons excited to the number of incident photons, is described...
Fig. 5. (a) The photoresponse performance of the Schottky photovoltaic PD at different bias. The response times of the Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky junction PD at (b) -4 V and (c) 4 V. (d) The variation of response speed with applied bias. Electron energy level diagrams of the device: (e) with applied reverse bias (f) with applied forward bias.

Fig. 6. (a) The rise and (b) decay times of the Schottky PD and conductive PD with different light intensities at 0 V and -0.1 V, respectively. (c) The $R$, EQE, and $D*$ of Schottky PD as a function of 254 nm illumination intensity at 0 V. (d) Wavelength-dependent responsivity of the Schottky PD at 0 V bias. (e) Time-dependent response of the photovoltaic PD measured at as-prepared and after 40 days at 0 V. (f) Responsivity and dark current of our device and other previously reported Ga$_2$O$_3$ Schottky self-powered PDs.
Schottky PD is a greatly promising device in the field of UV photodetectors. By comparing with Photoconductive PDs, Schottky PDs show superior performances at zero bias, mainly due to their high responsivity and selectivity. The responsivity is measured in the range from 5.6 mA/W at 255 nm to 15.5 mA/W at 380 nm, which indicates the excellent stability of the PD. The unchanged photocurrent for their practical application and commercialization, therefore it could be a good solar-blind UV-sensitive PD.

To further investigate the working wavelength range of the Schottky PD, the responsivity is measured in the range from 200 to 500 nm at zero bias. The responsivity of the device with a maximum value of ~5.6 mA/W at 255 nm is negligible to visible light, which closely matches the UV-vis absorption spectra in Fig. S3. For example, the UV-vis rejection ratio ($R_{\text{vis}}/R_{\text{400nm}}$) is found to be $2.9 \times 10^4$, confirming that Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky PD owns high wavelength selectivity with low background noise and could be a good solar-blind UV-sensitive PD.

The stability of PD is considered as a significant challenge for their practical application and commercialization, therefore it was investigated by testing the performance upon the exposure to air environment for 40 days under multi-cycle 254 nm UV illuminating at zero bias (Fig. 6e). The unchanged photocurrent indicates the excellent stability of the PD.

As shown in Fig. 6f, the Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky PD concomitantly shows high responsivity and low dark current, compared to the other self-powered Schottky junction Ga$_2$O$_3$ PDs. For a more comprehensive comparison, Table I lists the critical parameters of Schottky PDs based on the Ga$_2$O$_3$, including our work and other previously reported studies. Compared with other Ga$_2$O$_3$ Schottky PDs, our device shows higher responsivity, EQE, rejection ratio, lower dark current, moderate detectivity, and response speed. The superior performance indicates that our Ti$_3$C$_2$/ε-Ga$_2$O$_3$ Schottky PD is a great promising candidate for the fabrication of high responsivity and low noise PDs.

### References

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