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Near-ultraviolet photodetector based on hexagonal TmFeO$_3$ ferroelectric semiconductor thin film with photovoltaic and pyroelectric effects

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ABSTRACT

The switchable ferroelectric photovoltaic (FPV) effect facilitates application of multifunctional photoelectric devices. The drawback of the FPV effect is that it generates a very low photocurrent in highly insulated ferroelectric materials. In contrast, the light-induced pyroelectric effect enhances photoelectric performance. Both effects strongly depend on the ferroelectric polarization of the material. In this study, we fabricated and characterized a near-ultraviolet photodetector consisting of a Pt/hexagonal TmFeO$_3$/Pt heterojunction. The switchable FPV and light-induced pyroelectric effects are both observed in a hexagonal TmFeO$_3$ ferroelectric semiconductor film. An additional potential arises from the light-induced pyroelectric effect, which strongly depends on the light intensity. The Schottky barrier height can be modulated by both the poling electric field and light-induced pyroelectric potential. Increasing the power density above the threshold leads to switchable polarization via the light-induced pyroelectric potential. The coexistence of photovoltaic and pyroelectric effects in the hexagonal TmFeO$_3$ ferroelectric semiconductor makes it possible to develop electronic, thermal, and optical sensors as well as energy conversion devices.

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I. INTRODUCTION

Ferroelectric semiconductors have attracted much attention for developing electronic devices for information storage, signal transduction, and solar energy harvesting.$^{1-5}$ Properties such as ferroelectric and pyroelectric effects are highly useful for multifunctional applications.$^{6,7}$ In ferroelectric crystals, the remanent polarization holds two or more stable states in the absence of an external electric potential, and switching between these states can be achieved by applying an opposing electric field.$^8$ A switchable photovoltaic effect has been observed in ferroelectric semiconductors, in which the depolarization field leads to the separation of photogenerated electron–hole pairs.$^9,10$ The ferroelectric photovoltaic (FPV) effect often leads to unusual photoelectric characteristics in ferroelectric semiconductors. For example, semiconductors have exhibited anomalous photovoltages that exceed the bandgap.$^{11,12}$ The Schottky barrier commonly exists in photodetectors and plays a vital role in switchable FPV devices.$^{13,14}$ However, the drawback of FPV devices is that a very low photocurrent is generated in typical highly insulated ferroelectric materials. Some ferroelectric materials also exhibit pyroelectric properties whereby the polarization undergoes temperature-dependent changes resulting in a flow of charge (additional polarization). The light-induced pyroelectric effect generally results in ultrafast and enhanced photoelectric conversion, while pyroelectric polarization-related Schottky barriers lead to a high photocurrent.$^{15,16}$
As an alternative to increasing the power conversion efficiencies of conventional ferroelectric semiconductors, hexagonal rare-earth ferrites of the form RFeO$_3$ (R = Y, Dy, Lu) tend to have narrow direct bandgaps. Ferroelectricity has also been found in which spontaneous polarization occurs in the P6$_3$/cm phase at room temperature. A transition between the ferroelectric P6$_3$/cm and paraelectric P6$_3$/mmc phases occurs at the Curie temperature. Thus, solar cells have been fabricated using the FPV effect of hexagonal RFeO$_3$ films and have exhibited high solar absorption and stability during electrical fatigue tests. In addition, strain in thin films modulated via an appropriate substrate can reduce the bandgap and enhance polarization. This is due to changes in charge transfer between occupied O 2p orbitals and unfilled Fe 3d orbitals under the substrate-induced film strain. The interface potential between metallic electrodes and hexagonal RFeO$_3$ layers remains poorly understood. This interface plays an important role in the photovoltaic effect and can be adjusted between switchable polar states via an applied electric field.

Here, we report a near-ultraviolet photodetector based on a hexagonal TmFeO$_3$ ferroelectric semiconductor thin film. The switchable photovoltaic and light-induced pyroelectric effects are observed in the Schottky barrier between the Pt metal electrode and hexagonal TmFeO$_3$ (TFO) film. We study the migration of oxygen vacancies under a poling electric field and a light-pyroelectric potential. The photocurrent is investigated as a function of the incident light power density and poling electric field. The results reveal that the remanent polarization can be switched by the light-induced pyroelectric potential.

II. MATERIALS AND METHODS

The photodetector sample was designed in the Pt/TmFeO$_3$/Pt heterojunction-based structure shown in Fig. 1. Patterned Pt films were grown via magnetron sputtering on (0001)-oriented sapphire substrates as bottom electrodes. A 200-nm-thick hexagonal TmFeO$_3$ layer was deposited on the bottom Pt electrode via pulsed layer deposition (PLD) at a deposition temperature of 850°C and an oxygen pressure of 10 Pa. The laser frequency was 3 Hz, and the laser fluence was about 1.6 J/cm$^2$. Top Pt electrodes with a thickness of 5 nm were deposited via magnetron sputtering to avoid having an excessively thick Pt layer hindering the light absorption of the TmFeO$_3$. A 100-nm-thick Pt layer followed, serving as bonding pads for wire connection. Both the electrodes and pads were patterned using standard photolithography. The fabricated photodetector device was then fixed on a prepared printed circuit board (PCB), and copper wires were used to connect the top electrode pads and the PCB for photoelectric examination.

X-ray diffraction (XRD) patterns were recorded using a DX-2700BH multipurpose X-ray diffractometer at a scanning rate of 1.2° min$^{-1}$ in the 2θ range from 8° to 50° (Cu K$_\alpha$ radiation with a wavelength of 1.54056 Å). The surface morphology of the Al$_2$O$_3$/Pt/TmFeO$_3$ heterojunction was observed in noncontact mode using atomic force microscopy (AFM) with a Seiko SPA-300HV microscope. Absorption spectra were calculated through $\alpha = \ln(1/T)/t$, where $\alpha$ is the absorption coefficient, $t$ is the thickness of the hexagonal TmFeO$_3$ layer, and $R$ and $T$ are the reflectance and transmittance, respectively. Spectra were recorded using a Lambda 750 UV-vis/NIR spectrophotometer (Perkin-Elmer). Polarization was measured as a function of electric field, and both were recorded using a Precision Premier II ferroelectric tester (Radiant Technologies) to obtain their hysteresis loop.

We used a custom test platform controlled with the software LabVIEW (National Instruments) to measure the current density as a function of both voltage and time. Electrical curves were measured using a Model 6482 Dual-Channel Picoammeter/Voltage Source (Tektronix). The TmFeO$_3$ layer was illuminated with 405-nm light supplied by an ML-III Laser Generator (ChangChun New Industries Optoelectronics Tech. Co.) and controlled with a Model AFG1022 Arbitrary/Function Generator (Tektronix).

III. RESULTS AND DISCUSSION

Figure 2(a) is a schematic of the hexagonal TmFeO$_3$ (TFO) crystal structure with noncentrosymmetric P6$_3$/cm symmetry. Two distinct layers alternately stack along the c-axis: one characterized by a TmO$_8$ unit with trigonal D$_3$ symmetry and the other by a FeO$_8$ bipyramid with D$_{3h}$ symmetry. TFO films were deposited on Pt/sapphire (0001) substrates to prepare the photodetector to measure the photovoltaic effect. Only the (0002) and (0004) planes of hexagonal TFO were observed in the XRD pattern, as shown in Fig. 2(b). This suggests that TFO thin films grown using PLD preferentially exhibit c-axis orientation. The full width at half maximum (FWHM) of the diffraction peaks suggests an average particle size of $\sim 125$ nm according to the Scherrer equation. The AFM images in Fig. 2(c) show typical sphere-like grains in the hexagonal TFO film. The average grain size is about 100 nm, as shown in the inset of Fig. 2(c). The average root mean square (rms) roughness of the TFO film is 3.5 nm. Pinholes are randomly distributed in the film, which could suppress the ferroelectric performance of the TFO thin film. Room temperature Raman spectra were also taken to obtain better information on the TmFeO$_3$ sample, as shown in Fig. 2(d). The Raman peak positions and relative intensities are similar to those of hexagonal LuFeO$_3$ with P6$_3$/cm.
symmetry, which possesses six formula units per unit cell with 38 Raman-active phonon modes ($9A_1 + 14E_1 + 15E_2$). The peaks around 114, 271, 384, 459, 482, and 682 cm$^{-1}$ correspond to the $E_2$ modes, while the peak at $\sim$652 cm$^{-1}$ is assigned to the $A_1$ modes. The above observation reveals that the TFO sample has a polar hexagonal phase.

To examine the c-axis-oriented ferroelectric properties, the polarization-vs electric-field (P–E) loop was measured at room temperature, as shown in Fig. 3(a). The hexagonal TFO film exhibits a lossy dielectric loop rather than a perfect ferroelectric hysteresis loop. This indicates that the current conduction partly contributes to the mixed valence of Fe ions ($Fe^{2+}/Fe^{3+}$) or oxygen vacancies. The XPS spectrum of the Fe 2p region clearly indicates the $Fe^{3+}$ state, as shown in Fig. S1 of the supplementary material. Thus, oxygen vacancies might dominate the lossy behavior.

The bandgap of the TmFeO$_3$ sample was also determined from the Tauc plot, as shown in the inset of Fig. 3(b). All details for calculating $\alpha$ and the bandgap are included in Sec. II. The bandgap of the TFO sample was 1.98 eV, lower than the value of 2.09 eV reported by Hyeon Han et al. Meanwhile, the absorption coefficient does not smoothly rise below the wavelength of 620 nm corresponding to the onset of strong optical absorption. These phenomena can be typically attributed to defect engineering, where the oxygen vacancies described above narrow the bandgap via inserted defect levels. This can introduce recombination centers to change the absorption coefficient. Therefore, besides the intrinsic bandgap of 1.98 eV in hexagonal TFO films, defects such as oxygen vacancies are expected to have a non-negligible role in photoelectric conversion.

Figure 3(c) shows the current density vs voltage for the Pt/TmFeO$_3$/Pt heterojunction-based photodetector with upward and downward poling. Both the positive and negative branches show a nonlinear J–V response, indicating a Schottky barrier on the hexagonal TFO side of the Pt/TFO interface. The magnitude of the photocurrent at nonzero bias voltage obviously increases with increasing illumination intensity, which can be attributed to the photogenerated electron–hole pairs. However, the short-circuit photocurrent does not change significantly, implying that most of the photogenerated electron–hole pairs separate in the Schottky barriers. In addition, the J–V curves with the same light intensity change slightly when the poling field is applied before the test. This is related to the strength and direction, as shown in Fig. 3(d) and in Fig. S2(d) of the supplementary material. Therefore, the interfaces with Schottky barriers are modified by the applied poling field. Oxygen vacancies are also a major factor because they can migrate along the electric field and adjust the band structure via defect engineering. This can be verified by the difference in photocurrents between the two opposite scan directions for the bias voltage in the I–V test, as shown in Fig. S2 of the supplementary material.

To identify the photoelectric conversion, the dynamic photocurrent responses at zero bias voltage were measured by varying the poling electric field from $-0.75$ to $+0.75$ MV/cm$^2$, as shown in Fig. 4(a). Both the photovoltaic and light-induced pyroelectric...
effects are observed by analyzing the temporal response characteristics, as shown in Fig. 4(b). The corresponding current densities $J_{\text{pyro}+\text{photo}}$ and $J_{\text{photo}}$ are extracted as functions of the poling electric field strength, as shown in Fig. 4(c). An increase in the downward poling electric field significantly enhances the steady-state current density $J_{\text{photo}}$, while an increase in the upward poling electric field slightly decreases $J_{\text{photo}}$. The trend of the transient pyroelectric current density $J_{\text{pyro}}$ is similar to that of $J_{\text{photo}}$, implying that both the photovoltaic and light-induced pyroelectric effects involve the separation of electron–hole pairs. However, the orientations of $J_{\text{pyro}}$ and $J_{\text{photo}}$ cannot be switched by changing the polarization direction, despite that oxygen vacancies can be easily driven by the electric field. Electromigration of light-induced oxygen vacancies is evidently important in photoelectric conversion.

The unswitchable $J_{\text{photo}}$ and $J_{\text{pyro}}$ behavior indicates the prominent role of the light-induced pyroelectric effect on the electromigration of oxygen vacancies. Figure 4(c) shows the working mechanism of the light-induced pyroelectric effect in four stages. The sample is kept at room temperature without any illumination, and the charge distribution remains stable after a poling field is applied in stage I, where there is no net current generation. In the short moment after illumination turns on in stage II, the Schottky barrier induces a photocurrent and the incident light immediately increases the interfacial temperature by a few degrees Celsius. The light-induced temperature variation rate is positively correlated with light intensity. The incident light then induces pyroelectric polarization charges across the hexagonal TFO sample, with a transient pyroelectric potential from the top electrode to the bottom. After a few microseconds or even nanoseconds, the sample stays slightly above room temperature in stage III, where only a net photocurrent is output. When the illumination is removed in stage IV, the instantaneous temperature reduction also leads to a pyroelectric potential in the opposite direction to that in stage II. The amount of charge $Q_{\text{pyro}}$ driven by the pyroelectric effect is obtained by integrating the $J$–$t$ curve over the reference value $J_{\text{photo}}$. The time constant $\tau_{\text{Decay}}$ from the transient peak current $J_{\text{pyro}+\text{photo}}$ transiting to the steady-state current $J_{\text{photo}}$ represents the decay of the light-induced pyroelectric effect through the external circuit. The pyroelectric charge $Q_{\text{pyro}}$ and decay time constant $\tau_{\text{Decay}}$ are extracted and calculated as functions of the poling electric field, as shown in Fig. 4(d). The time constant $\tau_{\text{Decay}}$ is nonlinearly correlated with the pyroelectric charge $Q_{\text{pyro}}$, indicating a nonlinear change of resistivity in the hexagonal TFO film. This nonlinear resistivity is attributed to the variations of the oxygen vacancy distribution induced by the poling electric field. The pyroelectric charge $Q_{\text{pyro}}$ increases with increasing strength of the downward poling field, whereas the opposite trend occurs with an upward poling field. This indicates that the generation of $Q_{\text{pyro}}$ depends on the oxygen vacancy distribution, which is tunable using the poling electric field. The pyroelectric potential increases as the oxygen vacancies accumulate. The stronger...
pyroelectric potential enhances the concentration of photogenerated electron–hole pairs, leading to an increase in $\tau_{\text{Decay}}$. Therefore, the light-induced pyroelectric potential stimulates the electromigration of oxygen vacancies but also depends on the oxygen vacancy distribution.

To better understand the characteristics of the light-induced pyroelectric effect, the time-dependent $J_{\text{SC}}$ was measured with power densities from 2.56 to 22.96 mW/cm$^2$, as shown in Fig. 5(a). Figure 5(b) shows the extracted transient peak current $J_{\text{pyro+photo}}$ and steady-state current density $J_{\text{photo}}$ as functions of power density. As the power density increases, $J_{\text{pyro+photo}}$ and $J_{\text{photo}}$ increase for downward polarization, while the absolute values of $J_{\text{pyro+photo}}$ and $J_{\text{photo}}$ first steadily increase and then significantly decrease for upward polarization. A gradual increase in $J_{\text{pyro+photo}}$ occurs in the relatively high power density range of 15.2–22.96 mW/cm$^2$ when a light pulse is added. This also occurs in $J_{\text{photo}}$ to a lesser degree. Therefore, the continuous increase at high power densities ($\geq 15.2$ mW/cm$^2$) indicates that the Schottky barrier is largely modulated by the light-induced pyroelectric potential via pyroelectric-induced oxygen vacancy electromigration. The variations of $J_{\text{pyro+photo}}$ and $J_{\text{photo}}$ at low power densities ($\leq 12.8$ mW/cm$^2$) are similar to traditional behavior in which the response photocurrent is enhanced with increasing illumination intensity. \textsuperscript{42,43} The light-induced pyroelectric potential has a threshold power intensity of $\sim 15.2$ mW/cm$^2$ for overcoming the opposite depolarization field. This is exhibited as a reversal in the short-circuit current densities $J_{\text{UP}}$ and $J_{\text{dark}}$.

The light-induced pyroelectric effect is manifested as a combination of the drift and diffusion of hot carriers and the electromigration of charged objects such as oxygen vacancies. To confirm the light-induced pyroelectric effect on the Schottky barrier modulation, the corresponding photoresponsivity is calculated as $R = \frac{J_{\text{light}} - J_{\text{dark}}}{I_{\text{ill}}}$, where $J_{\text{light}}$ and $J_{\text{dark}}$ are the short-circuit current densities with and without illumination, respectively, and $I_{\text{ill}}$ is the power density. The transient photoresponsivity $R_{\text{pyro+photo}}$ in Fig. 5(c) significantly exceeds the steady-state photoresponsivity $R_{\text{photo}}$, significantly decreasing as the power density increases from 2.56 to 12.8 mW/cm$^2$. As the power density increases above the threshold of 15.2 mW/cm$^2$, $R_{\text{pyro+photo}}$ and $R_{\text{photo}}$ slightly increase for downward polarization. This indicates that the oxygen vacancy concentration in the Schottky barrier increases with the strong light-induced charge electromigration. For upward polarization, $R_{\text{pyro+photo}}$ and $R_{\text{photo}}$ significantly decrease with increasing power density in the low power density range and then increase slightly above the threshold power density. This implies that the photoelectric conversion is correlated with space charges in the Schottky barriers, which can be modified by the oxygen vacancy electromigration when the light-induced pyroelectric potential exceeds the threshold.

In general, the grain boundaries and pinholes were widely distributed in the films, which caused lattice distortion and the presence of irregular O$^{2-}$ sites in the vicinity.\textsuperscript{44,45} Most oxygen vacancies were trapped around the grain boundaries or pinholes and had enhanced mobility parallel to the boundary plane.\textsuperscript{46,47} Applying an external electric field concentrated the oxygen vacancies on one side of the grains, forming local built-in electric fields across the grain boundaries. Oxygen vacancies tend to pass through the grain boundaries when the total field strength exceeds a certain value. The concentration of oxygen vacancies increased on one side of the film even if the external electric field was removed, which led to changes in...
the band structure of the Schottky barrier. Energy band diagrams can be drawn for specific situations to reveal the physical mechanism of photoelectric conversion in the Pt/TFO/Pt heterojunction. Energy bands modulated by the migration of oxygen vacancies are shown in Fig. 6, which includes the effects of oxygen vacancies on the semiconductor properties. The polarization response to the migration and redistribution of oxygen vacancies arises from free ionic diffusion above room temperature. The Schottky-barrier-induced built-in potential and the depolarization field should both be taken into account for a ferroelectric semiconductor thin-film photodetector.

The poling electric field can trigger the electromigration of oxygen vacancies, leading to a layer with a high concentration of oxygen vacancies at the interface. This induces a heavily doped n+ layer, and the increase in the barrier height enhances the separation of electron–hole pairs, as shown in Figs. 6(a) and 6(d). The light-induced pyroelectric potential accumulates oxygen vacancies at the bottom of the hexagonal TFO film. Because the pyroelectric potential depends on the power density, the oxygen vacancy distribution does not exhibit obvious changes with light intensity below the threshold. Therefore, Fig. 5(b) shows a nearly linear dependence of the net short-circuit photocurrent on the power density, while the increasing poling electric field enhances the photocurrent response in Fig. 4(c).

IV. CONCLUSION

We observed the coexistence of the switchable FPV effect and light-induced pyroelectric effect in a hexagonal TmFeO$_3$ ferroelectric semiconductor thin film. This demonstrates that photoelectric conversion of a hexagonal TmFeO$_3$ film containing oxygen vacancies can be tuned by applying a poling electric field and a

FIG. 6. Band energy diagram of the Pt/TFO/Pt photodetector for upward poling (a) without light irradiation, (b) with a low illumination intensity, and (c) with a high illumination intensity. Band energy diagram for downward poling (d) without light irradiation, (e) with a low illumination intensity, and (f) with a high illumination intensity.
light-induced pyroelectric potential. A high oxygen vacancy concentration enhances the Schottky barrier, which largely provides the potential to separate electron–hole pairs. The light power density is a key factor for adjusting the distribution of defects via the light-generated temperature gradient. This provides a means to modulate the photoelectric conversion. These findings suggest the potential use of hexagonal TmFeO$_3$ ferroelectric semiconductor thin films in near-ultraviolet photocathodes, fast pyroelectric sensors, and energy conversion devices.

**SUPPLEMENTARY MATERIAL**

See the supplementary material for the high-resolution Fe 2p XPS spectra of the hexagonal TmFeO$_3$ film (Figure S1) and J-V characteristics of the Pt/hexagonal TmFeO$_3$/Pt photodetector (Figure S2).

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