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ABSTRACT

We report on the structural, chemical, and optical properties of titanium sesquioxide Ti_2O_3 thin films on single-crystal sapphire substrates by pulsed laser deposition. The thin film of Ti_2O_3 on sapphire exhibits light absorption of around 25%–45% in the wavelength range of 2–10 μm . Here, we design an infrared photodetector structure based on Ti_2O_3 , enhanced by a resonant metasurface, to improve its light absorption in mid-wave and long-wave infrared windows. We show that light absorption in the mid-wave infrared window (wavelength 3–5 μm) in the active Ti_2O_3 layer can be significantly enhanced from 30%–40% to more than 80% utilizing a thin resonant metasurface made of low-loss silicon, facilitating efficient scattering in the active layer. Furthermore, we compare the absorptance of the Ti_2O_3 layer with that of conventional semiconductors, such as InSb, InAs, and HgCdTe, operating in the infrared range with a wavelength of 2–10 μm and demonstrate that the absorption in the Ti_2O_3 film is significantly higher than in these conventional semiconductors due to the narrow-bandgap characteristics of Ti_2O_3 . The proposed designs can be used to tailor the wavelengths of photodetection across the near- and mid-infrared ranges.

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Infrared photodetectors are critical components of various advanced technologies designed to detect and respond to infrared light.^{1–6} Infrared photodetectors are devices designed to detect and convert infrared radiation into measurable electrical signals by utilizing materials and structures sensitive to wavelengths in the infrared spectrum. Conventional semiconductors used in infrared photodetectors include indium antimonide (InSb), indium arsenide (InAs), and mercury cadmium telluride (HgCdTe or MCT).^{7–9} These materials are chosen for their suitable bandgaps, allowing them to absorb infrared photons efficiently in Earth's atmospheric windows, including two wavelength regions: mid-wave infrared windows (MWIR: 3–5 μm) and long-wave infrared windows (LWIR: 8–12 μm). The detection mechanism typically involves excitation of electrons from the valence band to the conduction band upon absorbing infrared photons, generating an electrical signal.¹ Narrowband detection, a desirable feature in these photodetectors, refers to their ability to selectively detect a narrow range of wavelengths, which enhances sensitivity and specificity in capturing desired spectral information. For example, HgCdTe is the commercially used compound material in the photodetectors of

Fourier transform infrared spectrometers or night vision cameras. However, these HgCdTe-based photodetectors or cameras need to be cooled to near liquid nitrogen temperatures of ~ 77 K to reduce the thermally excited noise. In addition, these narrow-bandgap semiconductors usually contain hazardous elements such as As, Hg, Cd, and Te.

Unlike conventional materials, titanium oxides have emerged as promising narrow-bandgap alternatives due to their environmental safety. Titanium dioxide, TiO_2 , is known to be nontoxic, widely available, cost-effective, and environmentally safe. This material has shown great promise in various areas, such as photovoltaics, visible-light photocatalysis, and biomaterials. Although significant research has been conducted on the wide-bandgap rutile form of TiO_2 , its 3.3 eV bandgap is too large. Recently, great effort has been focused on improving the catalytic efficiency of TiO_2 by enhancing light absorption through doping or reduction.¹⁰ For example, in reduced TiO_{2-x} , the isolated Ti^{3+} defect structure plays an important role in reducing the bandgap and increasing absorption in the visible range.¹⁰ However, in this case, the Ti^{3+} ions are created within the Ti^{4+} structure of TiO_2 , which significantly limits their effect. Recent research

efforts aim to develop a stable and pure Ti^{3+} ($3d^1$) containing system such as titanium sesquioxide (Ti_2O_3).^{11,12} Ti_2O_3 is a lesser-known titanium oxide with potential applications in photodetection and other electronic devices due to its unique optical and electronic properties.^{11–13} Bulk Ti_2O_3 exhibits an insulator–metal transition at ~ 450 K, but unlike VO_2 , it does not experience a structural phase transformation.^{14–16} Upon heating, Ti_2O_3 transforms from an insulating state to a metallic state, showing typical semiconductor behavior with an ultra-narrow bandgap at low temperatures.^{11,15} This ultra-narrow bandgap and uniaxial anisotropy make Ti_2O_3 a promising candidate for specialized applications in infrared photodetectors and optical coatings.^{16,17}

Metasurfaces can enhance scattering into the active layer of solar cells and photodetectors, improving their performance when used as a coating.^{6,18–20} These surfaces are designed to support Mie resonances, which can control light at the subwavelength scale and increase the efficiency of light confinement and harvesting.^{21–23} In periodic metasurfaces, the Rayleigh anomaly occurs when a diffraction order becomes grazing to the plane, strongly influencing light scattering and enhancing lattice resonances. By tuning the directionality of the scattering, resonant metasurfaces can more effectively channel light into active regions, boosting the overall efficiency of these devices.^{24,25} The coupled multipolar models explain the collective optical response of periodic metasurfaces by describing the interaction between multipolar excitations of nanoantennas mediated by the lattice geometry.²¹

Here, we present the engineered structure of an infrared photodetector utilizing Ti_2O_3 , with performance improvements achieved by integrating a resonant metasurface. Our findings indicate that the absorbance of the thin Ti_2O_3 layer increases significantly and almost reaches 100% due to the addition of the resonant metasurface, resulting in efficient scattering within the active layer. Furthermore, we demonstrate that the Ti_2O_3 -based photodetector exhibits superior absorbance compared to photodetectors based on other commonly used infrared materials, such as InSb, InAs, and HgCdTe.

Epitaxial Ti_2O_3 thin films are grown on c-plane sapphire substrates by pulsed laser deposition (PLD) with a KrF excimer laser (Lambda Physik LPX 305, 248 nm, 30 ns FWHM). Details of the growth procedure can be found in Ref. 13. The chamber pressure is kept at high vacuum ($<10^{-8}$ Torr; 25 °C) during deposition, which is necessary to prevent the growth of impurity phases, such as anatase TiO_2 and rutile TiO_2 . The laser beam is focused on a rotating Ti_2O_3 target (2.56 cm in diameter, Kurt Lesker) with a repetition rate of 5 Hz and laser fluence of 1.5 J/cm². The laser beam was rastered across the target surface in order to uniformly ablate the target surface while the target was rotated. For uniform film deposition, the substrate was also rotated during deposition at a speed of 20 rpm. The films are grown at a substrate temperature of 720 °C. After growth, the samples are kept at these temperatures for 30 min and then cooled at a rate of 10 °C down to room temperature in a $\sim 10^{-8}$ Torr vacuum.

The crystal structure of the films is characterized by x-ray diffraction (XRD) measurements with Cu $K\alpha_1$ radiation (Bruker D8). Figure 1(a) shows a typical XRD pattern of a 200-nm-thick Ti_2O_3 thin film grown on an Al_2O_3 substrate at 720 °C. Peaks at $\sim 41.68^\circ$ and $\sim 90.70^\circ$ are indexed to the planes (0006) and (000,12) of the Al_2O_3 substrate, respectively. One can observe the film peaks at 37.9° and 81.0° , which correspond to the planes (011) and (022) of the orthorhombic phase ($\text{O-Ti}_2\text{O}_3$) (*Immm* space group).¹² However, as shown in Fig. 1(b), when we zoom in 2θ angles around the (022) peak, a weak

secondary peak is observed at $\sim 79.8^\circ$, which is indexed as (111) TiO phase, indicating that the film grows mainly in orientation (011) of Ti_2O_3 with a secondary phase of TiO . The areal ratio of $\text{Ti}_2\text{O}_3/\text{TiO}$ appears to be ~ 0.9 . Similar mixed phases of Ti_2O_3 and TiO are consistent with the previous report on Ti_2O_3 films grown by PLD.²⁶

The chemical structure of the grown films is characterized by Raman spectroscopy measurements (Witec, alpha 300) with a magnification of 100X and a laser wavelength of 532 nm. As shown in Fig. 1(c), four Raman peaks are observed at 247, 333, 446, and 612 cm^{-1} , which are assigned to orthorhombic Ti_2O_3 .¹² These Raman spectra also confirm the orthorhombic Ti_2O_3 phase determined by XRD measurements. However, the TiO phase is not observed in Raman spectroscopy measurement due to the relatively small amount of TiO compared to Ti_2O_3 . The surface morphology of the films is characterized using an Atomic Force Microscope (AFM, Bruker Dimension Icon) operated in peak force mode [Fig. 1(d)]. The film is composed of irregular shapes of large grains with a characteristic dimension of ~ 100 nm with rounded edges, and the film exhibits a relatively low surface roughness of ~ 2.3 nm.

The surface chemistry analysis of the films is performed using Al $K\alpha$ x-ray photoelectron spectroscopy (XPS) measurements. In order to remove contamination and eliminate surface oxidation effects, a high-energy argon (Ar) ion beam sputtering is applied for 210 s. Figure 1(e) shows the survey XPS spectra of the Ti_2O_3 film. As expected, before Ar-sputtering, a strong carbon peak is observed, and the ratio of O/Ti is close to Ti^{4+} (TiO_2 structure) due to surface oxidation. However, after Ar-sputtering, the carbon composition is reduced from 64 to 1 at. %, and the element ratio of O/Ti clearly decreases from 1.94 to 1.38 due to an increase in Ti^{3+} (Ti_2O_3 structure). As shown in Fig. 1(f), high-resolution XPS spectra of the Ti 2p region after Ar-sputtering verifies dominant Ti^{3+} states in Ti_2O_3 films.

The optical dielectric permittivity of the Ti_2O_3 films is determined by ellipsometry measurements by fitting the ellipsometry parameters **W** and **D** into the Drude oscillator models. Spectroscopic ellipsometry is performed using a Variable-Angle Spectroscopic Ellipsometer (VASE, J.A. Woollam Co., Inc.) for wavelengths from 1.7 to 10 μm . The ellipsometry and reflectance data of the Ti_2O_3 films are analyzed using WVASE software. Figure 2(a) shows the real and imaginary parts of the dielectric permittivity for Ti_2O_3 thin films grown on the (0006) Al_2O_3 substrate.

A continuous 200-nm-thick film of Ti_2O_3 on top of the sapphire substrate absorbs approximately 25%–45% of incident light in the wavelength range of around 2–10 μm [Fig. 2(b)]. The further increase in absorbance is limited by the high reflection at the interface of Ti_2O_3 and free space, which have drastically different permittivities. Nanostructuring of the top layer can help reduce the reflectance and increase the amount of light propagating through the layer, but it is challenging to accomplish and not as effective as covering the layer with the metasurface, which efficiently scatters light into the active layer. The resonant metasurface made of material with negligible optical losses, such as silicon, can help channel more light into the active photodetector section and increase its efficiency, which we analyze below, showing that the absorbance of the thin Ti_2O_3 layer can reach almost 100%.

When designing the hybrid photodetector structure, it is crucial to evaluate and select appropriate theoretical models to predict and optimize the absorption spectra effectively. Various approaches can

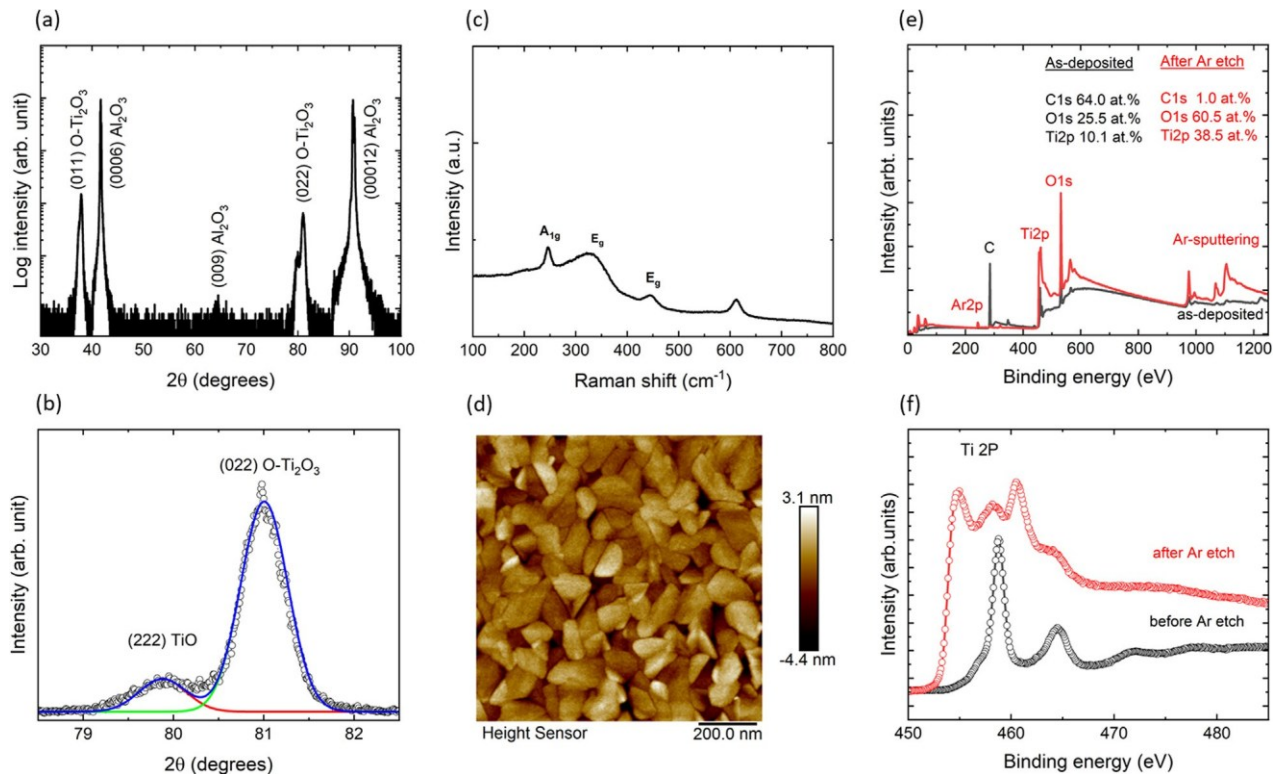


FIG. 1. XRD 2θ -x scans of Ti_2O_3 film grown on (0006) Al_2O_3 substrate in (a) a wide 2θ angle scan of 30° – 100° and (b) a zoomed 2θ angle scan of 79° – 82° . (c) Raman spectra of the Ti_2O_3 films. (d) AFM image of the film surface showing uniform surface morphology with grain sizes of approximately ~ 100 nm. (e) Survey XPS spectrum of the Ti_2O_3 film before and after Ar ion etching. (f) High-resolution XPS spectra of the Ti 2p region before and after Ar etching. The etching time was 210 s.

guide this process, including impedance matching theory, resonant critical coupling theory, and others tailored to specific photodetector configurations, mainly describing how resonant modes interfere in the structure.^{27–29} In regular periodic lattices composed of nanoantennas supporting Mie resonances, collective resonances emerge due to the

coherent coupling between the localized Mie modes of individual nanoantennas and the diffracted orders of the lattice. These resonances are strongly influenced by the Rayleigh anomaly, which occurs when diffracted order transitions from evanescent to propagating at specific wavelengths, leading to enhanced in-plane scattering. The coupled

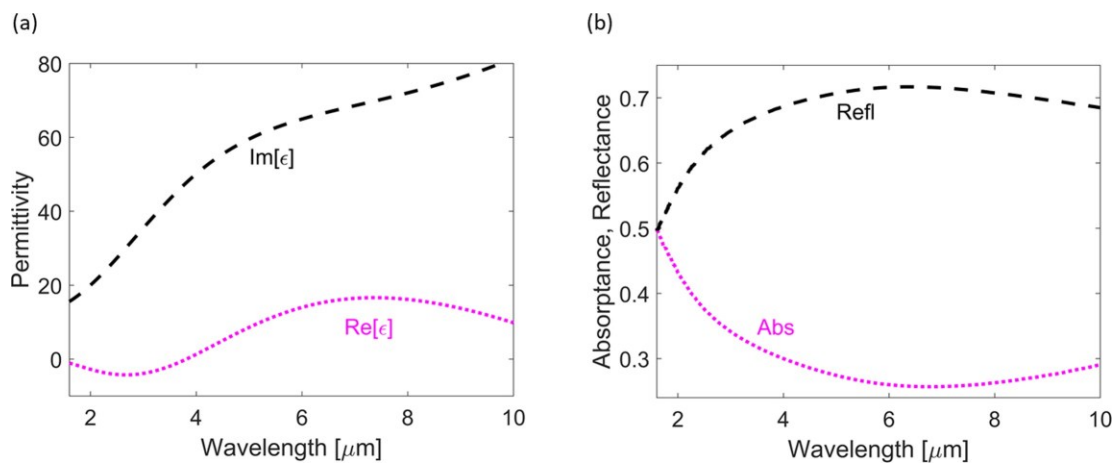


FIG. 2. (a) Permittivity ϵ of the ~ 200 -nm-thick Ti_2O_3 film grown on (0006) Al_2O_3 substrate by PLD. (b) Absorbance and reflectance of the same film on a sapphire substrate.

dipole–dipole model accurately describes these phenomena by considering the dipolar interactions between nanoantennas mediated through the lattice's periodicity in finite and infinite arrays.^{21,30} This model captures the hybridization of localized Mie resonances with lattice modes, resulting in sharp spectral features and increased quality factors characteristic of lattice resonances. Considering these diverse models enables a comprehensive exploration of the interplay between material properties, geometric parameters, and optical responses, ultimately leading to more robust and efficient photodetector designs.

Let us consider the following design of the infrared photodetector consisting of an active layer of Ti_2O_3 and a silicon metasurface, enhancing light concentration in the active layer [Fig. 3(a)]. The thickness of the active layer (made of Ti_2O_3) is t , and it is grown on a sapphire substrate, which is considered infinite in modeling. The height of the silicon nanoantennas is $a_z \approx 400$ nm, and it is fixed throughout the work. The nanoantennas have a square cross section with a_x and a_y , and they are arranged in a periodic array with d_x and d_y . Light is

linearly polarized along the x -direction and is normally incident on the device surface. Rayleigh anomalies can be observed in the spectra of the nanoantenna array at wavelength when the first diffraction order emerges. The wavelength corresponding to the (1,0) Rayleigh anomaly, where (1,0) denotes the first diffraction order in the x -direction and no diffraction in the y -direction, is defined as $k_{\text{RA}} \approx \pi/d_x$, where n is the refractive index of the material where first diffraction order propagates, and d is the period of the array in the x - or y -direction (assumed equal throughout this work). Silicon nanoantennas are selected because of their high refractive index and low material losses in the infrared range.

To analyze the effect of the silicon metasurface, we first fix the period of the nanoantenna array at $d_x \approx d_y \approx 3$ μm and the thickness of the Ti_2O_3 layer at $t \approx 200$ nm, and we vary the size of the silicon nanoantenna $a_x \approx a_y$ [Fig. 3(b)]. We observe that the absorptance enhancement occurs mainly near the Rayleigh anomalies. As the nanoantennas are in free space, the effect of the Rayleigh anomaly at

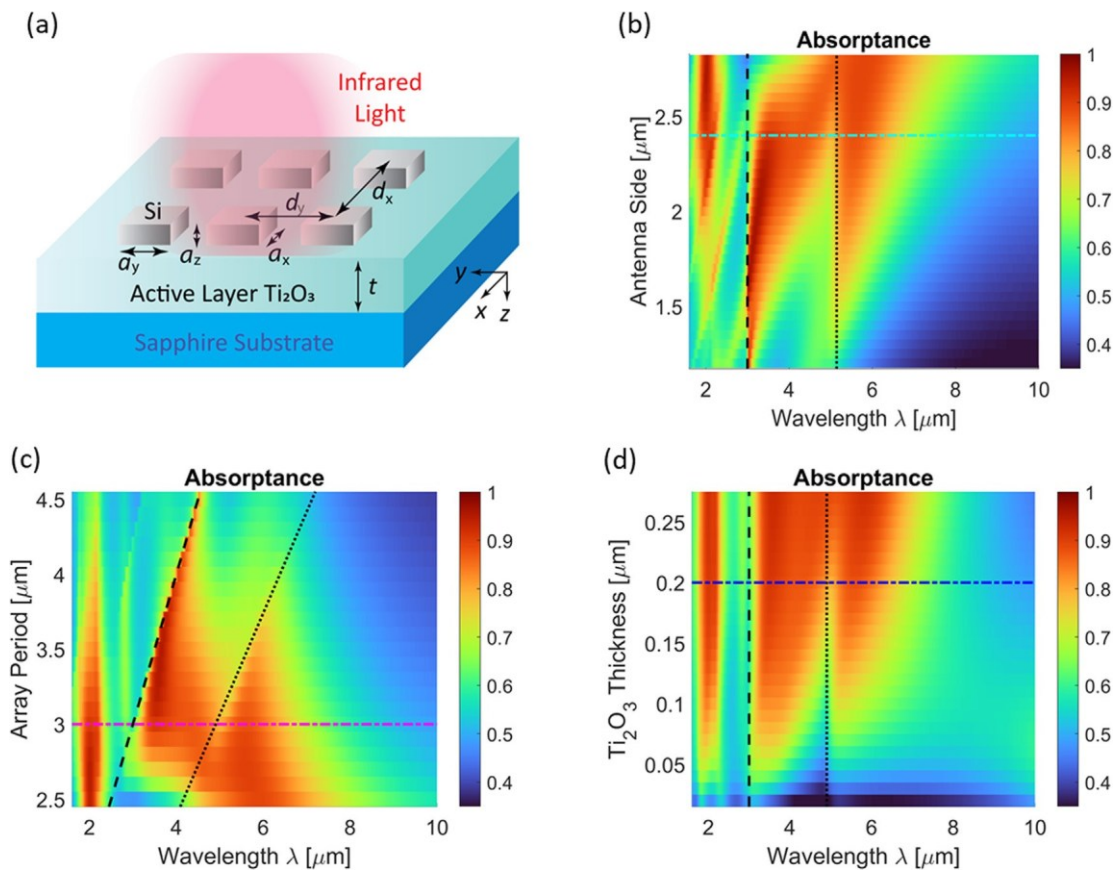


FIG. 3. (a) Schematic of the hybrid structure Si metasurface/ Ti_2O_3 / Al_2O_3 for enhanced absorptance in the infrared range. The top metasurface made of low-loss resonant nanoantennas (silicon) improves the scattering in the active Ti_2O_3 layer. (b) Absorptance of the nanoantenna array with a varying size of the silicon nanoantenna $a_x \approx a_y$. The array period is fixed at $d_x \approx d_y \approx 3$ μm , and the vertical dashed and dotted lines correspond to Rayleigh anomalies (1,0) in the free space and the sapphire substrate, respectively. The horizontal dotted-dashed cyan line corresponds to $a_x \approx a_y \approx 2.4$ μm , providing optimal performance and is selected for the analysis of other parameters of the metasurface. (c) Absorptance of the nanoantenna array for a varying period. The diagonal dashed and dotted lines correspond to Rayleigh anomalies (1,0) in the free space and the sapphire substrate, respectively. The horizontal magenta dotted-dashed line corresponds to $d_x \approx d_y \approx 3$ μm , providing optimal performance, and is selected for analysis of other parameters of the metasurface. (d) Effect of varying the thickness of Ti_2O_3 . Vertical dashed and dotted lines correspond to Rayleigh anomalies (1,0) in the free space and the sapphire substrate, respectively. The horizontal blue dotted-dashed line corresponds to $t \approx 0.2$ μm , providing the optimal performance, and is selected for the analysis of other parameters of the metasurface.

$k_{RA} \sim 3 \text{ } \mu\text{m}$ is stronger than the effect of the Rayleigh anomaly associated with the substrate. In the spectral region below the diffraction limit, the scattering in the active layer is not as effective as above the diffraction limit, and absorbance enhancement is observed only in narrow bands. For subsequent analysis, we select silicon nanoantennas with side lengths of $a_x \sim a_y \sim 2.4 \text{ } \mu\text{m}$, which results in a higher absorbance of the active layer and, therefore, is expected to provide optimal photodetector performance.

The period of the nanoantenna array substantially affects the photodetector properties because the collective resonance of the nanoantennas can be spectrally tuned and shifted to a desired range. Our simulations show that a relatively dense array of nanoantennas is the most beneficial for increasing absorbance [Fig. 3(c)]. We observe the most efficient absorbance in the wavelength range of 3–6 μm . Efficient operation at the longer wavelength requires an increase in the array period and a simultaneous increase in the nanoantenna size. Furthermore, absorbance can be enhanced by utilizing a thicker Ti_2O_3 film, provided that its quality is not affected by the growth process and the accumulation of defects. The simulations show that the thickness of around 200 nm results in an almost complete absorbance of light. No further increase in thickness provides substantial enhancement [Fig. 3(d)]. Overall, the silicon metasurface with $d_x \sim d_y \sim 3 \text{ } \mu\text{m}$ and $a_x \sim a_y \sim 2.4 \text{ } \mu\text{m}$ and the thickness of Ti_2O_3 layer $t \sim 200 \text{ nm}$ can provide the maximum absorbance of the proposed structure, and therefore, it can result in the optimal photodetector operation in the mid-wave infrared range.

The spectral response of the resonant metasurface without the inclusion of Ti_2O_3 is shown in Fig. 3(d) (see the case where the thickness of the material is zero). While the overall absorbance is significantly low, distinct spectral features remain observable. These features are particularly noteworthy, as the spectral positions of the resonances align closely with those observed in the hybrid structure containing Ti_2O_3 . Thus, the hybrid structure retains the fundamental resonance characteristics of the metasurface without Ti_2O_3 , with comparable widths and spectral positions, demonstrating that including Ti_2O_3 primarily enhances absorbance without fundamentally altering the underlying resonance behavior.

Finally, we compare several other materials that are known to be used in mid-infrared absorbance (Fig. 4). In the calculations, the InSb and InAs permittivity data are taken from Ref. 31, and the $\text{Hg}_{0.69}\text{Cd}_{0.31}\text{Te}$ permittivity data are taken from Ref. 32. We compare the cases with and without silicon metasurface (denoted “w/Si” and “w/o Si,” respectively). The parameters of the silicon metasurface and the thickness of the active layer are the same for all materials, and they are set to $d_x \sim d_y \sim 3 \text{ } \mu\text{m}$, $a_x \sim a_y \sim 2.4 \text{ } \mu\text{m}$, and $t \sim 200 \text{ nm}$. We note that this comparison provides only absorbance of the active layer without accounting for the efficiency of carrier generation and other aspects of photodetection. Simulations show that the absorbance of Ti_2O_3 is much higher than that of other materials. This can be explained by the high material absorption and the metallic nature of Ti_2O_3 . A continuous 200-nm-thick film of InSb on top of the sapphire substrate absorbs around 6% of the incident light at a wavelength of 2 μm , and this absorbance decreases as the edge of the bandgap approaches zero at a wavelength of $\sim 7 \text{ } \mu\text{m}$. Similarly, InAs and HgCdTe absorb 8% and 12%, respectively, at 2- μm wavelength, and it drops at lower photon energies (reaching zero at ~ 3.5 and $\sim 4 \text{ } \mu\text{m}$, respectively). By patterning the Si metasurface on top of these

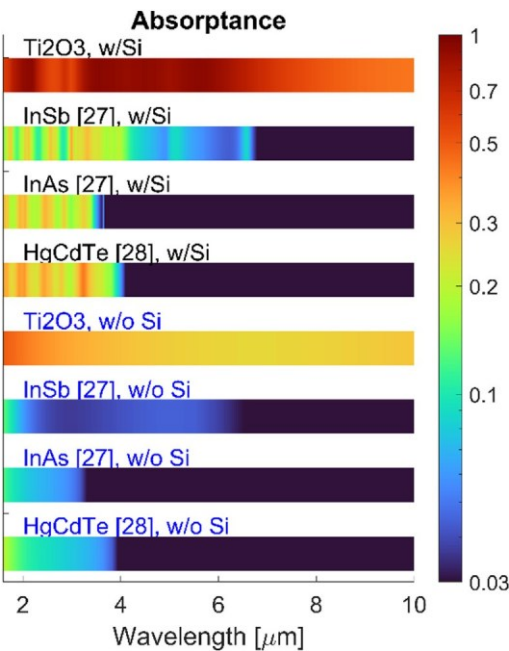


FIG. 4. Comparison of Ti_2O_3 with other materials commonly used in infrared photodetectors, such as InSb, InAs, and HgCdTe . The marks “w/Si” and “w/o Si” denote the case with and without the silicon metasurface, respectively. The parameters of the silicon metasurface and thickness of the active layer are the same for all cases.

conventional semiconductor layers, the absorbance of these active layers is slightly increased up to 8%–18% in the range of 3–5 μm . In contrast, the Ti_2O_3 layer without the Si metasurface absorbs $\sim 31\%$ in the mid-wave infrared window (3–5 μm), and by fabricating the Si metasurface, the average absorbance in the MWIR window increases up to 86%. The average absorbance in the range of 3–5 μm for each material with optimized metasurface parameters is summarized in Table I. Overall, the absorbance of the Ti_2O_3 layer is several times higher than that of InSb, InAs, and HgCdTe with the same Si metasurfaces due to the narrow-bandgap characteristics of Ti_2O_3 .

In conclusion, we have grown Ti_2O_3 thin films on a single crystal sapphire substrate by pulsed laser deposition. The 200-nm-thick Ti_2O_3 film on top of the sapphire substrate absorbs around 25%–45% of incident light in the wavelength range of around 2–10 μm . In order to

TABLE I. Average broadband absorbance in the mid-wave infrared window (3–5 μm) for Ti_2O_3 and other conventional semiconductors, such as InSb, InAs, and HgCdTe , with and without Si metasurface. The silicon metasurface with $d_x \sim d_y \sim 3 \text{ } \mu\text{m}$ and $a_x \sim a_y \sim 2.4 \text{ } \mu\text{m}$ and the active layer of $t \sim 200 \text{ nm}$ is considered in all cases. Labels “w/Si” and “w/o Si” denote the case with and without Si metasurface, respectively.

Active layer	Absorbance w/o Si	Absorbance w/Si
InSb	4%	18%
InAs	1.5%	8%
HgCdTe	4%	16%
Ti_2O_3	31%	86%

enhance light absorption in the Ti_2O_3 layer, we introduced a resonant metasurface on top of the Ti_2O_3 layer. Adding a resonant metasurface composed of a low-loss material significantly enhances absorptance by promoting efficient scattering within the Ti_2O_3 layer. Comparatively, the Ti_2O_3 -based structure demonstrates superior absorptance in the infrared (2–10 μm) over conventional infrared semiconductors, such as InSb, InAs, and HgCdTe due to the narrow-bandgap characteristics of Ti_2O_3 , which make Ti_2O_3 promising light absorbers in infrared photodetection applications.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Viktoriia E. Babicheva: Conceptualization (equal); Formal analysis (equal); Investigation (equal); Writing – original draft (equal). Evgeniya Lock: Formal analysis (equal); Funding acquisition (equal); Writing – review & editing (equal). Heungsoo Kim: Conceptualization (equal); Investigation (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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