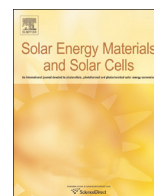




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## Highly efficient selective metamaterial absorber for high-temperature solar thermal energy harvesting



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### ABSTRACT

In this work, a selective solar absorber made of nanostructured titanium gratings deposited on an ultrathin MgF<sub>2</sub> spacer and a tungsten ground film is proposed and experimentally demonstrated. Normal absorptance of the fabricated solar absorber is characterized to be higher than 0.9 in the UV, visible and, near infrared (IR) regime, while the mid-IR emittance is around 0.2. The high broadband absorption in the solar spectrum is realized by the excitation of surface plasmon and magnetic polariton resonances, while the low mid-IR emittance is due to the highly reflective nature of the metallic components. Further directional and polarized reflectance measurements show wide-angle and polarization-insensitive high absorption within solar spectrum. Temperature-dependent spectroscopic characterization indicates that the optical properties barely change at elevated temperatures up to 350 °C. The solar-to-heat conversion efficiency with the fabricated metamaterial solar absorber is predicted to be 78% at 100 °C without optical concentration or 80% at 400 °C with 25 suns. The performance could be further improved with better fabrication processes and geometric optimization during metamaterial design. The strong spectral selectivity, favorable diffuse-like behavior, and good thermal stability make the metamaterial selective absorber promising for significantly enhancing solar thermal energy harvesting in various systems at mid to high temperatures.

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### 1. Introduction

Clean and abundant solar energy has been intensively explored as an alternative to traditional fossil fuels over the past decades [1]. As a key component, absorbers that convert solar radiation into thermal energy greatly affect the performance of various solar thermal systems. An ideal solar absorber should possess an absorptance of unity in the solar spectrum covering UV, visible and near infrared (NIR) to convert most solar radiation into heat, along with zero emittance in the mid-IR regime to minimize energy loss from spontaneous thermal radiation [2]. This spectral selectivity is vital for solar thermal absorbers to achieve high solar-to-heat conversion efficiency. In addition, angular and polarization independence is highly desired for efficient solar absorbers considering the random nature of solar radiation. Excellent thermal stability is also crucial for ensuring solar absorbers to operate properly and efficiently convert solar energy to heat at elevated temperatures

over time. Commercially, TiNOX [3] and Pyromark [4] have been used as solar absorbers for low- to high-temperature applications. Spectrally-selective TiNOX coatings could absorb 95% of incident solar radiation and emit only 4% thermal radiation, but its performance is optimal only around 100 °C. On the other hand, Pyromark exhibits a near-normal absorptance above 0.95 at high temperatures of around 650 °C, but its thermal emittance is also as high as 0.8. Unfortunately, efficient solar absorbers with both spectral selectivity and high-temperature compatibility are still lacking.

Optical metamaterials refer to artificial structures with exotic optical properties that cannot be obtained in naturally occurring materials [5]. Selective absorption has been investigated in metamaterials made of different micro/nanostructures from GHz to IR spectral regime, including split-ring-resonators [6,7], fishnet [8], cut-wires [9,10] and photonic crystals [11]. Recently, film-coupled metamaterials in metal-insulator-metal configurations have been intensively studied as thermal emitters or selective absorbers from visible to NIR spectral regime. Wang and Zhang designed a diffuse-like spectrally-selective thermophotovoltaic (TPV) emitter with 1D tungsten gratings on top of a SiO<sub>2</sub> spacer and tungsten substrate [12], while Zhao et al. modeled the

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polarization-independent 2D counterparts [13]. Note that selective TPV emitters can also be used as selective solar absorbers due to their spectral selectivity being well-matched to the solar spectrum. Photonic crystals [14,15] and nanoparticles [16–18] were also proposed for selective absorption in the visible and NIR range. Highly efficient solar absorbers require a broad absorption band from visible to NIR range, which could be possibly attained with lossy materials and geometric approaches. Wang and Wang proposed selective metamaterial solar absorbers with multi-sized tungsten patch arrays [19]. Aydin et al. discussed an ultra-thin broadband selective absorber composed of trapezoid gratings [20]. In addition, Lee et al. numerically explored a film-coupled nickel concave grating as a selective absorber for solar thermal energy harvesting [21].

Most of the previous experimental work on metamaterial absorbers has focused on optical characteristics at room temperature. Chen et al. demonstrated an IR selective absorber with film-coupled cross-bar structure at room temperature [22]. Hao et al. investigated a plasmonic NIR selective absorber but neglected the effects of temperature on material properties [23]. However, the temperature-dependent optical properties of novel metamaterial solar absorbers have to be characterized in order to clearly understand the temperature effect as well as thermal stability, which is crucial for ensuring efficient solar energy harvesting at elevated temperatures [24]. Yeng et al. measured the spectral radiance at high temperature for a selective emitter made of tungsten photonic crystal [25], but the temperature dependence of radiative properties like absorptance or emittance was not investigated experimentally. Liu et al. explored the emittance of a metamaterial thermal emitter at varied temperatures but only in the infrared range [26]. Recently, MIT researchers demonstrated a thermally stable selective solar absorber made of 2D metal-dielectric photonic crystal structures after 24 h heating at 1000 °C, but the temperature-dependent optical properties at high temperatures were not measured [27]. So far, measurements of the optical and radiative properties of selective solar absorbers at elevated temperatures have received little attention. The state of the art of the micro/nanostructured selective solar absorber was recently reviewed by Khodasevych et al. [28].

In this work, we report on the spectroscopic characterization at both room and elevated temperatures of a selective metamaterial solar absorber made of a 2D titanium grating deposited on an MgF<sub>2</sub> spacer and an opaque tungsten film, as illustrated in Fig. 1a. Tungsten is chosen as the substrate material due to its excellent high-temperature stability and high absorption in the visible and NIR regime. Titanium rather than tungsten in Ref. [19] is selected for the grating materials because of its easiness to pattern in deposition and lift-off processes. MgF<sub>2</sub> is used for the dielectric spacer considering its better CTE (i.e., coefficient of thermal expansion) match with tungsten and titanium than other dielectrics like SiO<sub>2</sub> to minimize the possibility of thermal cracking at high temperatures. The near-normal specular and hemispherical reflectance is measured over a broad spectral range from UV to the mid-IR regime, demonstrating its spectral selectivity. The electromagnetic field distribution obtained from finite-difference time-domain (FDTD) simulation is plotted at absorption peaks to explain underlying mechanism. The effects of oblique incidence, polarization state, as well as temperature up to 350 °C are further experimentally investigated. Finally, the solar-to-heat conversion efficiency for the metamaterial solar absorber is predicted to show its excellent performance especially at high absorber temperatures.

## 2. Sample fabrication

The selective metamaterial solar absorber is fabricated with the following procedure. Firstly, MgF<sub>2</sub> and tungsten thin films were deposited using e-beam evaporation (Kurt J. Lesker PVD 75) on a silicon substrate. Then, the 2D Ti gratings with period of 600 nm were fabricated onto the MgF<sub>2</sub>/W coated Si substrate using electron beam lithography by a multi-step exposure scheme on a FEI Nova Nano SEM with NPGS (J.C. Naby Lithography Systems, Nanometer Pattern Generation System), followed by e-beam evaporation and lift-off process. Fig. 1b shows the photo of the fabricated metamaterial solar absorber sample with a 5.4 mm by 5.4 mm pattern area on a 21 mm by 18 mm Si wafer. The fabricated grating patterns at the top layer of the metamaterial solar absorber have excellent symmetry in the *x* and *y* directions as seen from the top-view SEM image in Fig. 1c, while a trapezoid shape is observed from the side-view SEM image in Fig. 1d, which is typical for metallic gratings patterned from a lift-off process with negative photoresist. The measured geometric parameters are: grating period  $\Lambda=600$  nm, grating top width  $w_1=200$  nm, bottom width  $w_2=360$  nm, grating height  $h=170$  nm, and MgF<sub>2</sub> spacer thickness  $t=50$  nm. The tungsten layer has a thickness of 200 nm, which is opaque within the spectral region of interests.

## 3. Results and discussion

### 3.1. Spectrometric characterization of spectral-normal reflectance in UV to mid-IR regime

The specular reflectance of the fabricated solar absorber was measured by a Fourier Transform Infrared (FTIR) spectrometer (Thermo Fisher, iS50) along with a variable-angle reflectance accessory (Harrick Scientific, Seagull) at an incidence angle of 8° from 0.4 to 20 μm in wavelength with a spectral resolution of 4 cm<sup>-1</sup> in wavenumber. Due to the excellent geometric symmetry in the *x* and *y* directions of the sample, the spectrometric measurement was performed with unpolarized waves as the radiative property has negligible polarization-dependence at the near-normal direction. The reflectance from 0.4 to 1.1 μm in wavelength was measured by an internal Si detector, while an internal DTGS detector was employed at longer wavelengths beyond 1.1 μm. An aluminum mirror was used as the reference and the measured reflectance is normalized based on the theoretical reflectance of aluminum. The measured reflectance was averaged over three measurements (each with 32 scans) by interchanging the sample and reference to reduce the occasional errors during the measurement. In order to check the uncertainty of FTIR measurements, the reflectance of a reference Si sample (Virginia Semiconductor, Boron doped with resistivity of 60 Ω cm) was measured and compared with its theoretical value, showing the measurement uncertainty within 2%.

The directional-hemispherical reflectance  $R_{\text{d-h}}^{\circ}$  and diffuse reflectance were measured in a custom-built 8-inch integrating sphere (Labsphere, IS) at an incidence angle of 8°. An unpolarized monochromatic light from UV to NIR (i.e., 0.35–1.6 μm in wavelength) was provided by the tunable light source (Newport, TLS-250Q) with a spectral resolution of 10 nm. The light signal was modulated with an optical chopper and measured with a commercial Si detector (Thorlabs, SM05PD1A) and an InGaAs detector (Thorlabs, SM05PD5A) after lock-in amplification (Orion, Merlin). A silver mirror was employed as the reference and the measured reflectance is corrected with the theoretical reflectance of silver. The reflectance was averaged from five individual measurements. The hemispherical reflectance was measured without a light trap, while the diffuse reflectance was characterized with a light trap

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