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Ultra-broadband perfect solar absorber by an ultra-thin refractory titanium nitride meta-surface

Zhengqi Liu*, Guiqiang Liu, Zhenping Huang, Xiaoshan Liu, Guolan Fu

Jiangxi Key Laboratory of Nanomaterials and Sensors, College of Physics and Communication Electronics, Jiangxi Normal University, Nanchang 330022, Jiangxi, China

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ABSTRACT

Electromagnetic wave absorbers with thinner structural thickness but with broader spectral absorption bandwidth are more desirable for various applications in solar energy and optoelectronics. In this work, a refractory titanium nitride meta-surface with efficient, ultra-broadband solar energy absorption is theoretically designed and numerically demonstrated. The resulting 250-nm-thick meta-surface absorber exhibits an ultra-broadband perfect absorption over the whole ultraviolet-visible-near infrared range. With taking the $A > 90\%$ into account, the absorption bandwidth is up to 1110 nm with the wavelength range varied from 0.316 μm to 1.426 μm. The titanium nitride nano-resonators array and its coating structure of titanium dioxide array cooperatively provide multiple resonant modes, which therefore introduce strong coupling with the solar radiation and eventually produce an ultra-broadband absorption. The absorption spectrum can be feasibly manipulated via tuning the structural parameters. Most importantly, in sharp contrast to the common absorbers formed with metallic nano-resonators, the titanium nitride based solar absorber is with much stronger thermal stability, illustrating the impressive promise for wide applications such as thermo-photovoltaics and other high-power optoelectronic processes.

1. Introduction

Electromagnetic wave absorbers are the devices in which the illumination at the operating wavelength can be efficiently absorbed, and then transformed into ohmic heat or solar energy and other forms of energy. Electromagnetic wave absorbers can be dated back to 1902 by the observation of anomalous reflection dips of the metallic gratings due to the great effort made by Wood [1]. The reflection dip in the spectrum thereby produces a high absorption since the transmission is cancelled by the opaque metal substrate. In 2008, based on metal-insulator-metal triple-layer meta-materials, perfect electromagnetic wave absorbers were first reported by Landy [2]. Since then, numerous absorbers based on the meta-materials have been proposed and demonstrated from microwave to optical frequencies [3,4]. Nevertheless, the absorption bandwidth is often narrow, typically only a single absorption peak with respect to the central frequency by the plasmonic resonance. In many cases, broadband absorption is required, such as solar energy harvesting or photovoltaics, which are with the necessary to expand the absorption bandwidth and simultaneously to enhance the conversion efficiency [5]. Aiming at the purpose of broadening the absorption bandwidth, one may suggest using various different plasmonic nano-resonators together, which can provide corresponding absorption peak at the resonant wavelength [6,7]. However, the absorption bandwidth cannot be broadened significantly since the technique to compact different nano-resonators in a spatial unit cell is intrinsic limited. Recently, an average absorption of 71% has been demonstrated in the visible range (400–700 nm) by using a trapezoid metallic nano-resonators array, in which multiple plasmonic resonances can be excited due to the artificially designed size fluctuation of the resonant cell [8]. Based on the corrugated insulator-metal-insulator-metal architecture, high absorption up to 82.5% was reported in a broad spectral range from 300 to 750 nm [9]. In our previous study on the broadband light absorber, the average value of absorptivity was observed to reaching 83% across the spectral range of 370–880 nm [10]. Although broadband light absorption was obtained in these ultra-thin platforms, the absorption bandwidth is strongly hampered due to the limitation of finite sized nano-resonators. Recently, based on the using of sawtooth anisotropic meta-material slab, ultra-broadband light absorption was observed in the infrared region [11] due to the collective of a series of plasmonic resonant absorption bands by using tens of paired metal-dielectric nano-resonators. However, in energy harvesting or thermal emitting or hot electron generation applications [12–17], a suitable structural size scale especially the ultra-thin functional layer is extremely essential to improve the conversion efficiency [18].

* Corresponding author.
E-mail addresses: zliu@jxnu.edu.cn, lzhq86025@163.com (Z. Liu).

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Moreover, as for the common meta-materials based absorber, noble metals such as Ag and Au have been widely used as the plamsonic nanoparticles or the nano-resonators. Despite these noble materials can provide strong plasmonic resonances and enable the efficient conversion of solar light into electrons, which can only be happened with an energy well below the bandgap of the semiconductor but limited to the energy higher than the potential barrier. In addition, several realistic challenges remain for the using of noble metals including the high cost and low reserves, poor thermal (Au, Ag) and chemical (Ag) stability, complementary metal-oxide semiconductor (CMOS) incompatibility and diffusion into surrounding structures. These features inevitably hinder the practical applications of burgeoning solar energy, thermal photovoltaics, thermal emitting and hot electron generation, etc. Currently, aluminum (Al) nanoparticles and nanostructures have been proposed and demonstrated as alternative for plasmonic resonant material [19]. Perfect light absorption has also been observed by the using of Al plasmonic nano-resonators [20,21]. Besides the plasmonic resonant behaviors, Al is the third most abundant element on earth that is only one-thousandth of the cost of Au or Ag, suggesting a much lower cost for the potential applications [22]. Nevertheless, the low melting point, low thermal stability and poor chemical stability would hamper its potential application when high local temperature is generated under high power excitation such as laser or concentrated solar illumination [23].

In contrast to the plasmonic behaviors and the related perfect absorption of the meta-materials or plasmonic metals, strong electromagnetic resonant responses have also been observed and demonstrated in the dielectrics [24–26]. For instance, optical Mie resonances and electromagnetic induced transparency, and other plasmon-like responses have been observed in the high-index dielectrics such as Si, TiO_2, Ge and other semiconductors [27,28]. In our previous reports, multi-band and broadband light absorption have been obtained by the metal-dielectric nano-resonators composite system or the all-dielectric meta-materials [22,29]. Nevertheless, these dielectrics are usually with a limited absorption bandwidth. Moreover, these absorbers are with low thermal stability, indicating limitations for applications in high-power manipulation.

Recently, refractory material of TiN has been developed for resonant absorption. Even though the fundamental mechanical and optoelectronic properties of TiN have been studied so far from the experimental and theoretical points of view [30], very little is known about its plasmonic behavior. In these years, great efforts have been therefore made to explore its optical and electric features since the TiN is with a high melting point of 2930 °C and high temperature durability [31,32]. Owing to the intrinsic loss and intraband transitions near the Fermi level within the Ti d-band, strong plasmonic resonances can be excited in the optical region, which is the most impressive optical feature for this refractory material. Additionally, energetic or the hot electrons can be generated by the electron collisions with the TiN nanostructures or nano-resonators under the localized electric field or the local hot spots [33,34]. Based on a triple-layer TiN meta-material structure, broadband light absorption has been designed and demonstrated in the whole visible range of 400–800 nm, where strong thermal stability was observed, suggesting the promise for high temperature applications by the TiN nano-structures [35]. Broadband hot-electron collection for application in solar water splitting has also proposed and demonstrated by using the plasmonic response of the TiN [33]. Moreover, frequency selective thermal emitters and absorbers were also designed to show high thermal stability by the refractory material [36]. Although there were some interesting reports on the light absorption and its following applications in these years, the absorption bandwidth is still much narrower than that of the achieved ultra-broadband absorption by the conventional metallic systems [10,11]. Aiming at the solar energy harvesting and the corresponding thermal photovoltaics, it is essential to achieve broadband absorption with the bandwidth in proximity to that of the solar spectrum. Additionally, the ultra-thin material layer is also significant for applications on efficient photoelectronic effects, hot-electron generation and collection.

In this study, we take use of the refractory material of TiN to realize the ultra-broadband perfect solar absorber (UPSA) with a simple and straightforward method. The absorption is exceeding 90% over a continuous spectral bandwidth of 1110 nm in the ultraviolet-visible-infrared wavelength range (0.316–1.426 μm) in an ultra-thin structure, indicating a near-unity absorption in the main solar radiation range. TiN is an emerging functional material with plasmonic behaviors in the optical region [35,37]. Moreover, TiN can provide compatibility with CMOS technology, corrosion resistance and high-temperature resistance, as well as mechanical strength and durability. That is, TiN nano-resonators can outperform noble metals in terms of thermal and chemical stability, mechanical strength and durability. Otherwise, the coating TiO_2 nano-resonators in the UPSA cannot only protect the configuration of the TiN but also form an Ohmic junction with the TiN at the interface, which therefore allows "downhill" hot electron collection into the semiconductor and enabling higher conversion efficiencies [33]. Thereby, the proposed refractory titanium composite meta-surface can hold novel applications on the thermal hot electron generation in an ultra-broadband solar energy spectrum. Meanwhile, in contrast to the noble metals of Au and Ag or other metals such as W and Ta [38], titanium has a higher reserve. Under current science and technology, TiN could be synthesized and fabricated commonly via chemical vapor deposition and physical sputtering deposition methods [39–41]. These distinguished physical and optical properties make the absorber one of the most encouraging and impressive candidates for refractory optoelectronic devices.

2. UPSA with ultra-thin TiO_2-TiN composite nano-resonators meta-surface

The schematic of the UPSA is shown in Fig. 1a. It is composed of a periodic TiN disks array coated by a TiO_2 disks array with the same size, which are patterned on a 50-nm-thick SiO_2 film and the following a 100-nm-thick TiN film opaque substrate. TiN structure has been used to be the main contributor for the optical field coupling and absorption due to its plasmonic properties in the visible-infrared frequency range [35,42]. As for the TiN material, its intrinsic refractory property with a melting point as high as 3200 K can hold it with ultra-high structural stability for the applications on high-power and high-temperature operations [32]. TiO_2 is a widely used high-index dielectric, which was demonstrated with strong plasmon resonances [28]. Moreover, its semiconductor property hold wide applications on the photoelectrical effects and particularly in the photocatalytic activity under ultraviolet irradiation. Titanium has a relatively larger world reservation in comparison with other metals such as W and Ta [38]. As for this UPSA scheme, the experimental fabrication can be described by a three-step process. First, homogeneous TiN flat film with a certain thickness value can be synthesized on a clean substrate via chemical vapor deposition [39] or physical sputtering deposition [40]. Second, a 50-nm-thick SiO_2 film can be deposited onto the TiN flat film using magnetic sputtering. Flat TiN and TiO_2 layers with the corresponding film thicknesses can be then deposited onto the SiO_2 film with the same sputtering technique. Finally, the periodic patterned arrays of the TiO_2 and TiN nano-resonators can be fabricated via standard lithography procedures such as the electro-beam and/or interference lithography, laser writing and/or reactive ion beam etching [35,41–43].

In order to investigate the optical properties and absorption mechanism of the UPSA, a numerical simulation is performed by three-dimensional finite-difference time-domain method [44]. The unit cell of the TiN or TiO_2 nano-resonators array is designed with a period (P) of 300 nm, height (h) of 50 nm, diameter (D) of 220 nm. Periodic boundary conditions were adopted in both the x- and y-directions to reproduce the array pattern and to reduce the computational memory. Perfectly matched layers were employed in the z-direction immediately