

METAMATERIALS AS BROADBAND ABSORBERS, TUNABLE COLOR FILTERS, AND MULTI-FUNCTIONAL METASURFACES

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ELECTRICAL AND ELECTRONICS ENGINEERING

By
Majid Aalizadeh

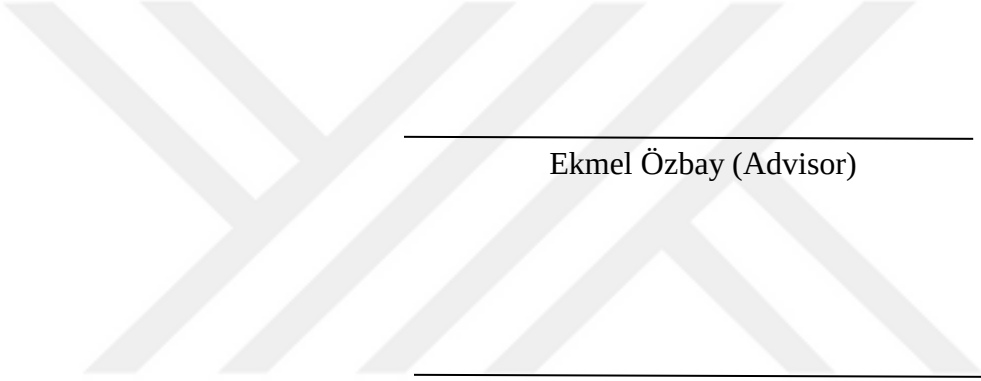
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By Majid Aalizadeh

June 2019

We certify that we have read this thesis and that in our opinion it is fully adequate, in scope and in quality, as a thesis for the degree of Master of Science.



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ABSTRACT

METAMATERIALS AS BROADBAND ABSORBERS, TUNABLE COLOR FILTERS, AND MULTI- FUNCTIONAL METASURFACES

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M.S. in Electrical and Electronics Engineering

Advisor: Ekmel Özbay

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Metamaterials have enabled us to come up with artificial structures and designs that can perform optical functionalities which are not achievable with natural materials. Here we design and implement three important applications of metamaterials as: 1. Ultra-broadband absorbers, 2. Real-time tunable color filters, and 3. Wideband and wide-angle efficient beam deflector and Multi-functional angular filter. By lithography being a major hinder on the way to mass production and cost-effectiveness, most of our works are lithography-free. We have introduced Manganese (Mn) for the first time as a very promising metal for broadband absorption and have used it in all our works. Four different Mn-based broadband absorbers are designed and fabricated in different chapters. Mn is used in the Metal-Insulator-Metal (MIM) cavity, annealed MIM configuration, top-layer-patterned MIM configuration, and random nanopyramids. It is shown in all works that Mn has a much better performance compared to other metals. For instance, in the work based on random nanopyramids, we obtain ultraviolet (UV) to far-infrared (FIR) perfect absorption by exploiting a lithography-free method and only by coating a single Mn layer on a high-roughness substrate. Moreover, using the combination of the MIM cavity and an electro-optic material, we have shown that a lithography-free color filter can be achieved that covers the whole visible spectrum by changing the voltage from -12 to 12 volts. Finally, in a structure composed of Silicon nano-rods, an ultra-wideband and wide-angle highly-efficient beam deflection is obtained. What makes the same structure very promising is that it

also has multifunctional applications as band-pass, band-stop, and low-pass angular filter.

Keywords: Broadband absorbers, Manganese, electrically tunable, beam deflection, and angular filtering.



ÖZET

GENİŞBANT EMİCİLER, AYARLANABİLİR RENK FİLTRELERİ, VE ÇOK İŞLEVİLİ METAYÜZEYLER OLARAK METAMALZEMELER

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Metamalzemeler, doğal malzemelerle elde edilemeyen optik işlevleri yerine getirebilecek yapay yapılar ve tasarımlar geliştirmemizi sağlamışlar. Burada şu üç önemli metamalzeme uygulama tasarlıyor ve uyguluyoruz: 1. Ultra geniş bant emiciler, 2. Gerçek zamanlı ayarlanabilir renk filtreleri ve 3. Geniş bant ve geniş açılı verimli ışın saptırıcı ve Çok fonksiyonlu açılal filtre. Litografi seri üretime ve maliyet etkinliğine giden yolda önemli bir engel teşkil ettiğı için çalışmalarımızın çoğı litografi içermemektedir. Manganez'i (Mn) ilk defa geniş bant emilimi için umut verici bir metal olarak tanıttık ve tüm çalışmalarımızda kullandık. Dört farklı Mn tabanlı geniş bant emici, farklı bölümlerde tasarlanmış ve üretilmiştir. Mn, Metal-İzolatör-Metal (MIM) yapısında, tavllanmış MIM konfigürasyonunda, üst katman desenli MIM konfigürasyonunda, ve rasgele nanopiramitlerde kullanılır. Tüm çalışmalarda Mn'nin diğer metallere göre çok daha iyi bir performansa sahip olduğu gösterilmiştir. Örneğın, rastgele nanopiramitlere dayanan çalışmada, litografi içermeyen bir yöntemden faydalanarak ve sadece yüksek pürüzlü bir alt tabaka üzerinde tek bir Mn tabakasını kaplayarak ultra-morötesi (UV) ile infrared (FIR) mükemmel emilimini elde ediyoruz. Ayrıca, MIM yapısının ve bir elektro-optik malzemenin kombinasyonunu kullanarak, voltajı -12 ila 12 volt arasında değıştirerek, tüm görünür spektrumu kaplayan litografi içermeyen bir renk filtresinin elde edilebileceğini gösterdik. Son olarak, Silikon nano çubuklardan oluşan bir yapıda, ultra geniş bant ve geniş açılı yüksek verimli ışın sapması elde edilir. Aynı yapıyı ümit verici kılan şey aynı zaman-

da bant geiři, bant durdurma ve dūřük geiřli aısal filtre gibi ok iřlevli uygulamalara sahip olmasdır.

Anahtar sōzcükler: Geniř bant emiciler, Manganez, elektrikle ayarlanabilen, kiriř sapması, ve aısal filtreleme.



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Chapter 1

Introduction

1.1 Background

This chapter is on part reprinted with permission from: 1. **Majid Aalizadeh**, A. Khavasi, B. Butun, and E. Ozbay, “Large-Area, Cost-Effective, Ultra-Broadband Perfect Absorber Utilizing Manganese in Metal-Insulator-Metal Structure,” *Scientific Reports*, vol. 8, p. 9162, 2018, and 2. **Majid Aalizadeh**, A. E. Serebryannikov, A. Khavasi, G. A. E. Vandenbosch, and E. Ozbay, “Toward Electrically Tunable, Lithography-Free, Ultra-Thin Color Filters Covering the Whole Visible Spectrum,” *Scientific Reports*, vol. 8, p. 11316, 2018.

Natural materials have limitations in their optical performance. They either have to be too bulky to be used for optical applications, or do not perform as efficient as required. Metamaterials, consisting of natural materials but in specific designs, have made it possible to come up with subwavelength structures that can function much more efficient compared to the natural materials.

In this thesis, three important applications of metamaterials are explored: 1. Ultra-broadband and lithography-free absorbers, 2. Lithography-free electrically tunable color filters, and 3. Wideband and wide-angle beam deflector and multifunctional angular filter metasurfaces.

1.1.1 Broadband Absorbers

Electromagnetic (EM) wave absorbers have attracted a lot of interest in technology, because of their vast range of applications in photovoltaics [1-4], shielding [5, 6], thermal imaging [7-9], sensing [10-12], photodetection [13-17], and thermal emission-based cooling [18, 19]. With the advent of plasmonics [20] and metamaterials, it became possible to design absorbers with ultra-thin sub-wavelength thicknesses, which is of high importance for optical packed circuit integration and the reduction of mass and cost of structures.

The general requirements for an absorber structure to absorb electro-magnetic (EM) energy are having impedance matching with free space to minimize reflection [21, 22] as well as near to zero transmission. One of methods to absorb light is to trap EM wave in a resonance mode of light and to absorb it by means of loss mechanisms such as electron collisions. The common resonance modes used in the design of absorbers are Localized Surface Plasmon (LSP) modes [23-27], Propagating Surface Plasmon (PSP) modes [28, 29] and Fabry-Perot resonance modes [30, 31]. To obtain broadband absorption, we can broaden the resonance bandwidth by sufficiently lowering the quality-factor with the appropriate choice of metals and the geometric design. The other approach is to design the structure in a way to have the superposition of several adjacent resonance modes and, therefore, broaden the absorption spectrum.

It is of high importance for the broadband absorber to be designed in a way to have large-scale production compatibility. One big barrier to large-scale production is the use of lithography in the structures, which is time consuming and costly. In order to have mass production compatibility, it is also very beneficial to use inexpensive materials in the structure. Therefore, designing a broadband lithography-free absorber in which inexpensive materials are used is very advantageous and meets the requirements of large-scale production.

In one of the pioneer works that was based on the superposition of LSPs, Aydin et al. [32] achieved an average of 71 percent absorption in the visible region (400-700 nm), by superposing the LSP modes of varying width of nanorods. However, it was achieved by use of Electron Beam Lithography (EBL). Some other works are fabri-

cated or designed to obtain broadband absorption in the visible [33-35], near-infrared (NIR) [36, 37], mid-infrared (MIR) [38-41], far-Infrared (FIR) [42], and Terahertz [43-46] ranges by using EBL and implementing the above-mentioned ideas.

In a recent lithography-free work conducted in our group, 97 percent absorption was achieved from 400 to 2000 nm wavelength by employing the superposition of LSP modes of dielectric-metal core-shell nanowires. This broadband perfect absorption was the result of randomness in the size of chemically synthesized nanowires [47]. One other common method to broaden the absorption band without the use of lithography is to anneal the structure in high temperatures, which is called the De-wetting process [48]. This leads thin layers to reform into nanoholes or nanoparticles, and adds the superposition of the LSP modes of nanopatterns into the absorption bandwidth. Some works are done in our group to broaden the absorption of Metal-Insulator-Metal-Insulator (MIMI) structure using this method [49, 50]. However, this comes with the cost of access to very high temperatures, which is time consuming and always brings the possibility of sample cracking, especially for the ones with thin substrates.

As mentioned earlier, lowering the quality factor of Fabry-Perot resonance is another way to achieve broadband absorption without lithography. This resonance can be obtained by making a Fabry-Perot resonator using an MIM structure. By adding the number of Metal-Insulator (MI) pairs, a broader bandwidth can be obtained. Some lithography-free works have achieved broadband absorption by employing MI multi-layer stacks. These works are done by using different combinations of MI pairs such as Cr-SiO₂ [51], W-Al₂O₃ [52], Ti-SiO₂ [53], and W-Al₂O₃-Ti-Al₂O₃ [54]. Also in a recent lithography-free work, broadband infrared absorption is obtained by implementing ITO-photoresist pairs [55].

1.1.2 Tunable Color Filters

The electromagnetic (EM) response of an optical device depends on the dimensions and material parameters of its components. This often imposes a limitation on their tuning functionality in a way that the EM response gets determined at the stage of

fabrication, since new dimensions and/or materials are required to obtain a new EM response. To obtain the possibility of real-time tuning of the EM response of optical structures after the fabrication, dynamically tunable ideas are required to be implemented. Among different ways of tuning, tuning with variable voltage (electrical tuning) has always been the most desirable and has been the topic of extensive research over the last decade. For this purpose, different methods and materials have been suggested. Utilizing electro-optically active materials is one of the most common methods for electrical tuning of optical structures. Optical behavior of these materials is modified by applying voltage. Liquid crystals [56-59], organic crystals [60-62], graphene [63-66], heavily doped compounds like Al-doped ZnO (AZO) [67] or n-type Indium Antimonide (n-InSb) [68], and transparent conductive oxides like Indium Tin Oxide (ITO) [69-74] or Indium Zinc Oxide (IZO) [75] are common materials used for this purpose. The choice of an electro-optic (EO) material depends on the design and the operating wavelength range of the optical structure. It is noteworthy that micro-electro-mechanical systems (MEMS) or nano-electro-mechanical systems (NEMS) have also been used to provide optical devices with the possibility of real-time controlling [76, 77].

Recently, several works have been reported, in which ITO is used to electrically tune the reflection spectrum of the optical structures, in particular, the one of the reflect-array metasurfaces [69-74]. Some works have been dedicated to the use of ITO as the tunable material to actively steer the reflected beam of metasurfaces [73, 74]. The structures involving ITO as the active material generally use it in the configuration of Metal-Oxide-Semiconductor (MOS) capacitor, so that by applying voltage, the carrier concentration of ITO changes in the vicinity of the ITO-oxide interface. These changes lead to those in the plasma frequency of the Drude model of the permittivity of ITO, and thus modulate the permittivity and the refractive index profile of ITO [78]. This is how the optical response of such structures gets tuned by the application of voltage.

A number of works have also used graphene as the active material [63-66]. The tuning in these works has been possible due to the shift in the Fermi energy level of graphene, which is obtainable by applying a bias voltage, so that the optical response of graphene and the whole structure is changed.

Color generators represent a wide class of the optical devices having various applications including displays, printing technologies, imaging, and light emitters. Some structural color generators are available in nature in specific types of insects, birds, and animals, such as butterfly wings [79-83]. Initial artificial color displays have employed liquid crystals, and produce the three main colors, red, green, and blue [84-86]. Recently, metamaterials have been introduced as a new platform that enables the realization of color displays with higher compactness, better resolution, better power-efficiency, and more flexibility in design-based color tunability. There are a lot of works demonstrating color filters without the possibility of real-time tuning [31, 87-93]. At the same time, tunable color filters working in the transmission or reflection mode have also been reported; some of which are dynamically controllable by the polarization of incident light [94-96], while some others are electrically tunable [56, 59, 68, 97]. For example, it has been suggested to use a polarization-tailored dichroic resonator combined with a twisted nematic liquid crystal [98]. However, in this case, lithography is required in the fabrication process.

In most of the earlier works, lithography is typically required as a part of fabrication process, which is the main barrier to mass production. It brings many limitations such as significantly restricting the area of samples, decreasing the throughput, and increasing the cost of fabrication. Therefore, coming up with a lithography-free design is of great importance for the purpose of large-area production and cost reduction. A lithography-free approach has been recently proposed in a few works for color filters, which cannot be tuned in real time [73, 74]. These series of lithography-free bandpass filters are based on the MIM cavities and work in the transmission mode.

1.1.3 Beam Deflectors and Angular Filters

Electromagnetic beam manipulation has attracted a lot of interest in recent decades. Beam deflection and anomalous reflection is one important type of beam manipulation application for wave-front engineering devices. The previous structures used for beam deflection include photonic crystals, hyperbolic metamaterials, gratings [99, 100], phased arrays, and metasurfaces [101, 102]. However, most of them either have a limited efficiency or do not have wideband and wide-angle desired behavior.

Another highly-demanded type of beam manipulation is the filtering functionality. Filtering can be realized both in the frequency and incidence angle domain, which are called spectral and spatial filtering, respectively. Spatial filtering is the possibility of passing or rejecting beams with a specific direction of incidence into another specified direction. These direction selective spatial filters (SFs) along with the spectral filters are of high importance in optics and have applications in detection, sensing, and imaging.

Some spatial filters were reported previously which were based on photonic crystals [103-105], resonant and non-resonant grating structures [106, 107], axisymmetric photonic microstructures [108], hyperbolic metamaterials [109], and waveguide-based systems [110]. Nevertheless, they are typically designed to have single functionality and often perform as one type of SF, i.e. either function as band-pass, band-stop, low-pass, or high-pass filters.

1.2 Thesis Outline

Chapters 2 to 5 are dedicated to four different broadband absorbers with the inclusion of Mn. Mn is introduced as a promising metal for broadband absorbers for the first time and it is shown in all works that it demonstrates a superior performance compared to other metals. In chapter 2, Mn is used in the basic MIM configuration and by using this lithography-free structure, above 94% average absorption is obtained in the wavelength range of 400-900 nm. Chapter 3 shows how annealing the Mn-based MIM structure boosts the broadband absorption performance. It is shown that by annealing the structure, random nanoholes appear in the top thin Mn layer that can support Localized Surface Plasmon modes, the superposition of which leads to broadening the absorption spectrum of the MIM cavity with a 2.1-fold enhancement. An average absorption of above 94% is achieved in the range of 450-1400 nm, using this lithography-free structure. In another attempt in chapter 4, it is shown that embedding an ultra-thin Mn layer inside the top-layer-patterned MIM cavity enhances the absorption band at about tenfold. As the last work on broadband absorbers, UV to FIR

absorption is achieved by coating a single layer of Mn on any high-roughness substrate and the results are presented in chapter 5.

In chapter 6, by exploiting an electro-optic dielectric in the insulator section of the MIM filter, we have achieved a lithography-free color filter that covers the whole visible spectrum by applying -12 to 12 volts of voltage.

Chapter 7 shows that by using a simple structure consisting of Silicon nanorods, highly-efficient, wideband and wide-angle beam deflection is achievable. This metasurface also shows multiple angular filtering functionalities as band-pass, band-stop, and low-pass spatial filters.

Finally, chapter 8 presents a conclusion on the thesis.

Chapter 2

Manganese-Based MIM Cavity

This chapter is on part reprinted with permission from: **Majid Aalizadeh**, A. Khavasi, B. Butun, and E. Ozbay, “Large-Area, Cost-Effective, Ultra-Broadband Perfect Absorber Utilizing Manganese in Metal-Insulator-Metal Structure,” *Scientific Reports*, vol. 8, p. 9162, 2018.

2.1 Introduction

It is known well that the MI-stack broadband absorbers have at least four layers (two MI pairs) in their structure. However, the purpose of this section is not to design an MI-stack absorber. We want to design a broadband absorber using the tri-layer MIM structure, which is the simplest possible configuration for absorbers. Moreover, as mentioned before, the MI-stack absorbers are discussed in several works, but, to the best of our knowledge, there are very few tri-layer broadband, lithography-free and annealing-free absorbers, reported previously [111].

In the present section, we experimentally demonstrate lithography-free, ultra-broadband nearly perfect absorption within 400-900 nm range by the MIM structure. The measured average absorption in this wavelength range is above 94 percent by the use of Mn in the metal layers. The most important difference between our work in this section and earlier works is that we introduce Mn for the first time in broadband

absorbers, and, by detailed analysis, we show that it is a very promising metal for achieving maximum absorption in broadband absorption applications. It is also an inexpensive metal unlike gold or silver, which can reduce the cost of fabrication in favor of mass production. Also, the proposed structure does not need post-annealing.

In the following parts of this section, first, MIM cavities and formulation of their resonance modes is discussed. Then, the optimization process of this structure is explained in detail in terms of two factors: 1. Materials and 2. Dimensions. First, to optimize the structure by choosing the appropriate materials, the effect of dielectric material is investigated, and also it has been demonstrated numerically that the best choice of metal is Mn for this structure. During the optimizations and calculations, the structure is modeled by using the Transfer Matrix Method (TMM) and also circuit theory. Some of simulations are also performed by the Finite Difference Time Domain (FDTD) method. It is also conceptually demonstrated as to how Mn is such an appropriate metal for the MIM structure and for broadband absorbers in general, by discussing based on its permittivity profile. Then, the structure is optimized in terms of the dimensions of different layers. At the end, the optimal structure has been fabricated and characterized. The experimental results are in good agreement with simulations and calculations showing a very large absorption bandwidth.

2.2 Design and Theory

Figure 2.1(a) depicts a 3D graphical schematic of the MIM structure and Fig. 2.1(b) shows its 2D cross section in xz plane. The bottom layer is an optically thick metal layer that is much thicker than the skin depth of the metal to ensure zero transmission. The middle insulator layer can be chosen among different dielectrics, taking into account the fact that the refractive index of the dielectric is an important parameter in determining the resonance frequency of the cavity mode. The top layer must be optically thin to allow the coupling of the incident light into the MIM cavity and consequently getting trapped inside it with back and forth reflections from metals. The trapped light is partly absorbed in each reflection, eventually, leading to complete absorption.

The resonance mode of the asymmetric Fabry-Perot cavity shaped by this MIM structure follows the equation of the form [30]:

$$2\left(\frac{2\pi}{\lambda_{res}}\right)n_id_i + \varphi_b + \varphi_t = 2\pi m \quad (2.1)$$

where λ_{res} is the resonance wavelength, n_i and d_i are the refractive index and thickness of the insulator layer, respectively, and m is an integer number which determines the order of cavity mode. It can be deduced from equation (2.1) that higher order cavity modes resonate at smaller wavelengths. φ_b and φ_t are the phase shift acquired from reflection from the bottom and top metal layers, respectively. It is noteworthy that equation (2.1) along with all other analytical theories presented throughout this section is only applicable for normal incidence. It can be seen that by increasing n_i or d_i , λ_{res} increases as well. In other words, by increasing the optical beam path which is written as n_id_i , the resonance wavelength red-shifts. Therefore, to modify the center of the absorption band, one can use dielectrics with different refractive indices or adjust its thickness according to the absorption band of desire. To achieve broadband absorption, the quality-factor of cavity mode should be sufficiently low, which is essentially obtained by the correct choice of metal.

To calculate absorption (A), reflection (R) and transmission (T) must be calculated and then absorption can be obtained from the $A=1-R-T$ formula. In the present section, TMM is used for these calculations, and the results are verified by FDTD simulations and circuit theory calculations. It should be noted that, for a sufficiently thick bottom metal layer, T is almost zero and, therefore, the absorption is simplified to $A=1-R$. To be more precise, however, we include T in our calculations.

2.2.1 Material Optimizations

In this part, the effect of dielectric material and the metal is investigated and the optimized materials are chosen among them.

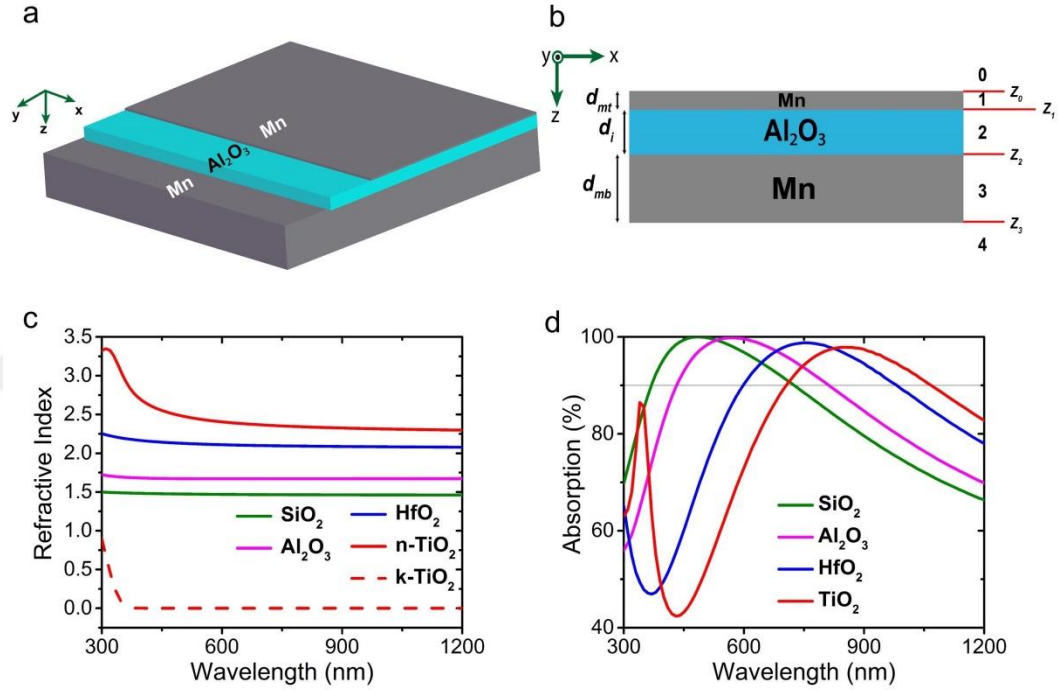


Figure 2.1: (a) 3D schematic of the proposed MIM structure, (b) cross section of the structure in xz plane, (c) refractive index of four common dielectrics investigated in the absorption calculations and, (d) calculated absorption spectrum of MIM structure.

Fig. 2.1 (c) shows the refractive index of four different common dielectrics: SiO_2 , Al_2O_3 , HfO_2 and TiO_2 , and Fig. 2.1 (d) shows the calculated absorption spectrum of MIM using the four mentioned dielectrics, while keeping the metal (Mn) and all dimensions constant. The thickness of the layers from bottom to top is 200, 65, and 5 nm, respectively. As expected, the absorption mode red-shifts as the refractive index of the dielectric material increases. Since we have the possibility to deposit Al_2O_3 by Atomic Layer Deposition (ALD) which deposits layers with lower roughness, the dielectric material is chosen to be Al_2O_3 for the rest of this work. It has to be mentioned that based on previous explanations and as it will be demonstrated in detail in the following sections, this thickness of the insulator layer, 65 nm, has been chosen by using Al_2O_3 to obtain high absorption in the desired bandwidth of 400-900 nm. It should be noted that other dielectrics can also be used, considering the fact that in order to achieve high absorption in the same wavelength band, the thickness of the dielectric layer should be calculated accordingly and the optimized thickness for this layer will be different from 65 nm.

In order to choose the most appropriate metal, as it will be elevated in detail in the next parts, the input impedance of the structure should be calculated. Therefore, first the calculation of the input impedance is explained. The input impedance of the proposed three-layer structure is calculated by circuit theory.

The equivalent circuit of the structure is shown in Fig. 2.2 (a). At normal incidence, β_m and Z_m are the propagation constant and characteristic impedance (normalized to the impedance of free space) of the transmission line corresponding to the metal layer and are equal to $n_m\beta_0$ and $1/n_m$, respectively, with n_m denoting the refractive index of the metal and β_0 denoting the propagation constant of light in vacuum. β_i and Z_i are similarly the corresponding parameters for the transmission line modeling the insulator layer. Finally, Z_0 is the normalized impedance of free space, which is equal to 1. It is assumed in this model that both the top and bottom metal layers have the same material, therefore, for both layers, the characteristic parameters are denoted with the subscript of ‘ m ’.

To better understand the mechanism of the absorption, we simplify the equivalent circuit by modeling the top thin metal layer with a surface conductivity, which is an accurate approximation for ultra-thin layers [39]. The characteristic equivalent admittance (surface conductivity) of the top layer (normalized to free space admittance) is

$$Y_{mt} = j(1 - \varepsilon_m)\omega d_{mt}/c \quad (2.2)$$

where c is the free space speed of light and ω is the angular frequency. ε_m denotes the relative permittivity of metal, and d_{mt} represents the top metal layer thickness. The above equation is obtained by approximating the bulk conductivity by surface conductivity when the layer is ultra-thin. Then the value of equivalent admittance of that layer is equal to the value of surface conductivity.

The second approximation is assuming the bottom metal layer thickness to be infinite. Therefore, Z_1 in Fig. 2.2(a) can be replaced by a load impedance of Z_m . Thus, the circuit model of the structure gets simplified into Fig. 2.2(b).

The input impedance seen from the interface of the top metal and the dielectric layer which is denoted as Z_2 in figures 2.2(a) and 2.2(b) can be written as [112]

$$Z_2 = Z_i \frac{Z_m - jZ_i \tan(\beta_i d_i)}{Z_i - jZ_m \tan(\beta_i d_i)} \quad (2.3)$$

In this approach, the admittance of top Mn layer or Y_{mt} is parallel to the equivalent input admittance of the other two layers which is equal to the inverse of input impedance of these two layers or Z_2 . The total input admittance then, is the sum of admittances Y_{mt} and Y_2 .

After calculating the input impedance (admittance) of the structure, the reflection of the structure, can be calculated by the following formula

$$R = \left(\frac{Z_{in} - 1}{Z_{in} + 1} \right)^2 \quad (2.4)$$

which is the well-known relation for reflection at the interface of two media with different impedances. Now, we approximate T to be zero, which is a very good approximation for a sufficiently thick bottom layer. We show the accuracy of this approximation by calculating transmission in the next sections. Therefore, the absorption can be simply calculated as $A=1-R$.

Now, for choosing the appropriate metal, we assume that the top and bottom metal layers both are chosen as the same material. Then we try to find the best metal for the structure. The material of metal layers plays the main role in determining the quality factor of the cavity. For this purpose, we calculate the normalized input impedance of the structure using circuit theory and then we equate it to the impedance of free space (i.e. 1). This is the condition for having zero reflection. From there the permittivity of the ideal metal can be calculated. Thereafter, the metal with the closest permittivity profile to the ideal metal should be chosen. During this calculation, the dimensions are 5, 65, and 200 nm, respectively and Al_2O_3 has been used as the insulator layer.

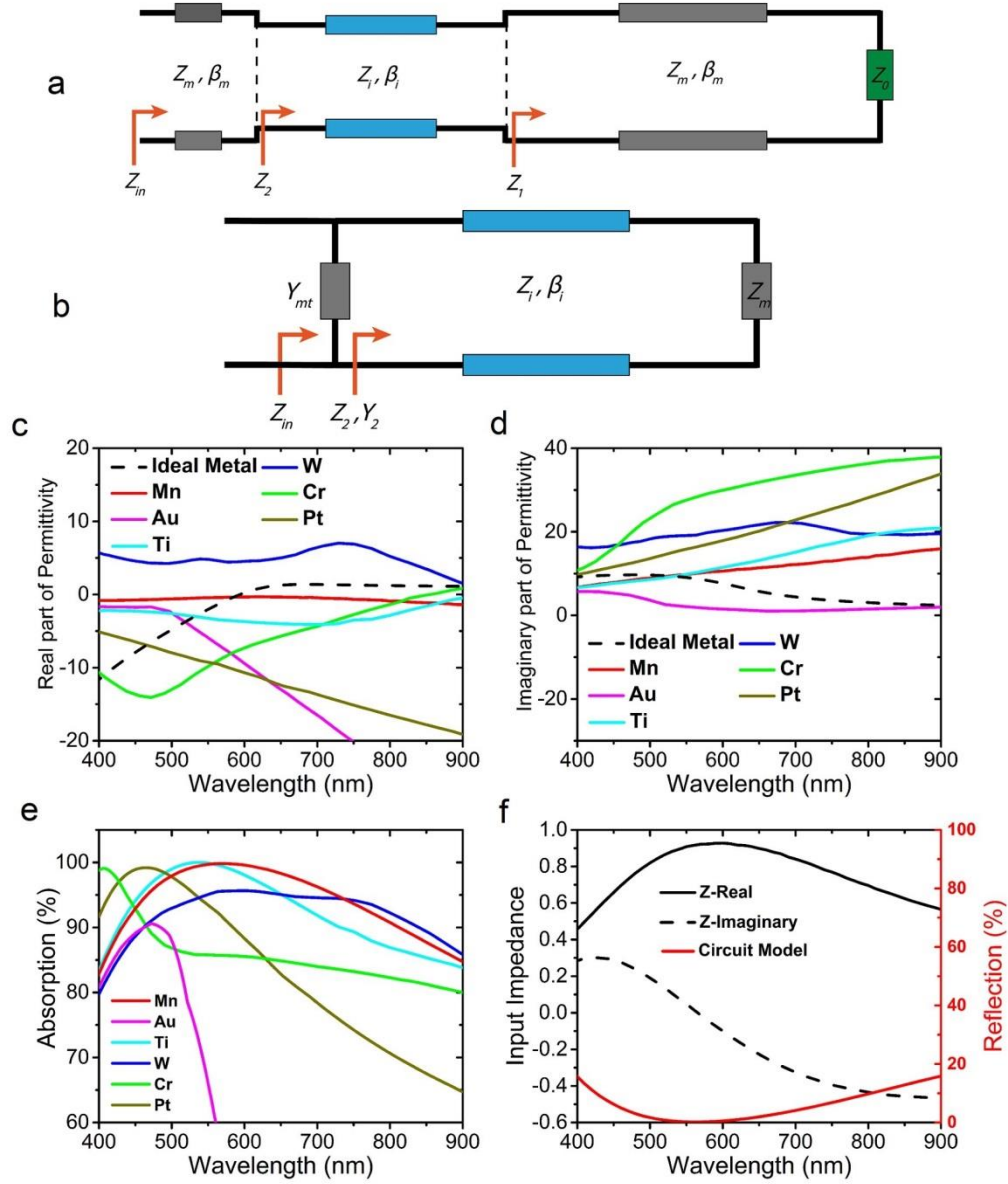


Figure 2.2: (a) Equivalent circuit of the MIM structure, (b) simplified equivalent circuit model of the MIM structure, (c) real and (d) imaginary part of permittivity of Mn and the ideal metal, along with those of Ti, W, Cr, Pt, and Au that are commonly reported metals for broadband absorption, (e) calculated absorption spectrum of the MIM structure using metals mentioned in (c) and (d). (f) Real and imaginary part of input impedance of the structure using Mn (black solid and dashed lines), normalized to free-space impedance, and reflection calculated using circuit theory (red line).

The calculated real and imaginary parts of relative permittivity profile of the ideal metal as well as some other metals are shown in Fig. 2.2 (c) and Fig. 2.2 (d), respectively. The metals are Titanium (Ti), Tungsten (W), Platinum (Pt), Chromium (Cr), and Gold (Au). Apart from Au, other metals are the candidates reported so far for broadband absorption applications, especially for lithography-free structures. Au can be used for broadband absorption in the case of patterning. The permittivities of the above-mentioned metals are plotted in these figures along with Mn, for a fine comparison between their permittivity profile and the one for ideal metal. The permittivity of the ideal metal is shown by black dashed lines in figures 2.2 (c) and (d). As it is obvious from both figures 2.2 (c) and 2.2(d), Mn has the closest permittivity profile to the ideal metal. It can be observed that the imaginary part of permittivity of the ideal metal is very close to that of Mn, from 400 nm up to around 600 nm. On the other hand, after 600 nm, up to the end of our desired spectrum, i.e., 900 nm, the real part of permittivity of Mn is similar to the real part of permittivity of the ideal metal. This demonstrates that Mn is the best candidate for our structure in our bandwidth of interest. So we choose Mn for the rest of our work as the top and bottom layer metal. It can be observed that around the calculated absorption peak wavelength, both the real and imaginary parts of permittivity of Mn almost intersect with the one for ideal metal. The material data of Mn and Al_2O_3 shown here, and also used in all the simulations and calculations, have been obtained by fitting spectroscopic data obtained from ellipsometry measurements. Material data for all other metals except for Au are obtained from Palik and for Au the data are obtained from Johnson and Christy.

Figure 2.2 (e) shows calculated absorption when the 5 different mentioned metals are used, along with the case of using Mn. It is obviously notable that, as predicted, Mn has the best absorption in terms of absorption strength and bandwidth at the same time. This is due to the fact that using Mn provides a better and broader impedance matching to free-space that leads to the minimization of reflection and trapping of light in a broad spectrum. Figure 2.2 (f) shows the real and imaginary parts of the calculated input impedance of the structure normalized to that of free-space, as well as reflection calculated using equation (2.4). It can be seen from this figure that the reason of having a broad dip in the reflection spectrum (broad absorption peak) of this structure lies in the fact that the real part of input impedance is close to 1 in a broad

wavelength band, while the imaginary part of it is close to zero. This minimizes the reflection and provides a broadband impedance matching to the free space.

Figure 2.3 (a) shows the input admittance of the structure. To have admittance matching with free space, similar to impedance matching, the real part of input admittance must be close to 1 and its imaginary part must be close to zero, which is the case here as it can be seen from Fig. 2.3 (a). The admittance matching process reveals better why Mn is such an appropriate metal for this structure. The real and imaginary parts of the equivalent admittance of top Mn layer and the input admittance seen from the dielectric and the top metal interface which is denoted as Y_2 in Fig. 2.2 (b), are shown in Fig. 2.3 (b). It is obvious that the real part of Y_2 is around 0.5 within the wavelength range of interest. The other required 0.5 of admittance for admittance matching is provided with the admittance of top Mn layer, as it can be seen in the figure. Fig. 2.3 (a) shows that the real part of input admittance of the structure stays around 1, but its imaginary part does not have such a behavior and its absolute value grows by getting far from the resonance wavelength. This limits near-to-perfect absorption to around 400-900 nm. It can be seen from Fig. 2.3 (b) that the imaginary part of admittance of the top Mn layer is around zero and, therefore, has almost no contribution to admittance mismatch happening outside the bandwidth.

Figures 2.3 (c) and (d) show the real and imaginary part of relative permittivity of Mn along with previously mentioned other 5 metals in a broad wavelength range between 400 and 2000 nm. By taking a look at these figures one can understand more about the physical reason behind broadband absorptive behavior of Mn. Compared to other metals, Mn has a very slowly changing real part of permittivity in the broad range of 400-2000 nm, covering visible and NIR, which makes impedance matching of the structure to that of free space easy. Its real part of permittivity has small values in this broad spectrum, which leads to high field penetration; and, at the same time it has a large imaginary part of permittivity that leads to high absorption. The lossy behavior of Mn results in a low quality factor Fabry-Perot MIM cavity. On the other hand, some other metals such as Gold and Silver have a very narrowband MIM cavity mode, because they are low loss metals.

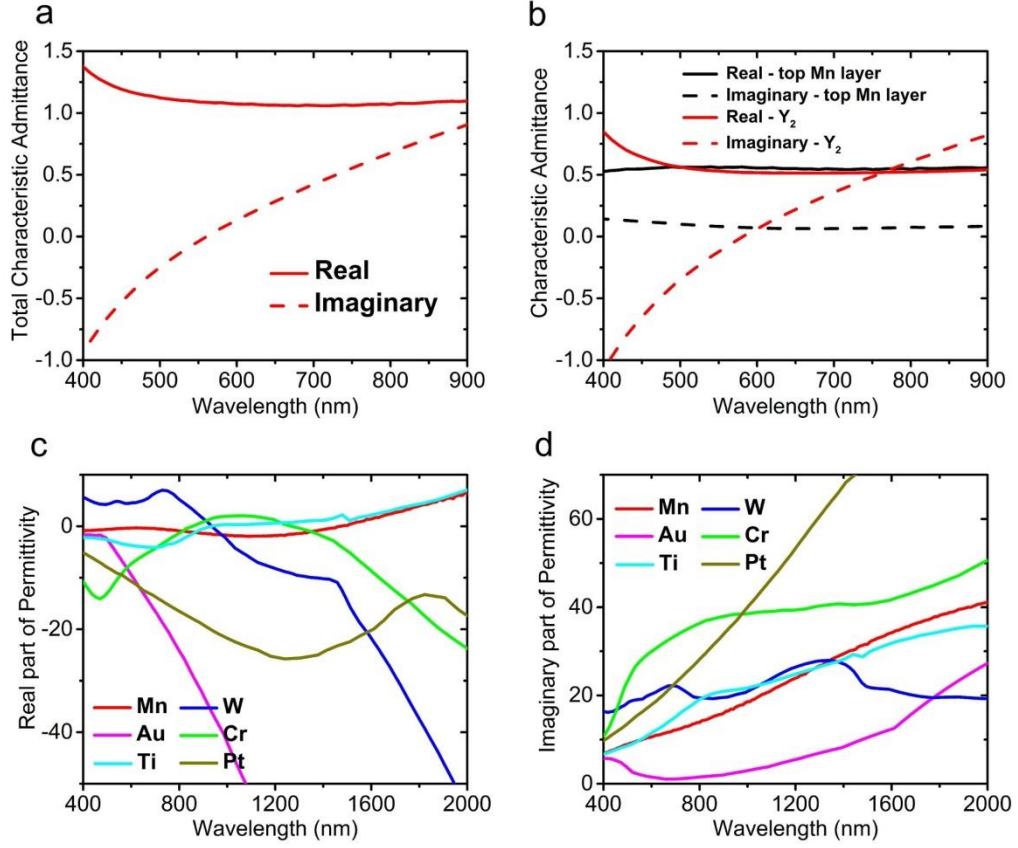


Figure 2.3: (a) Total characteristic admittance of the structure, calculated by circuit theory, (b) characteristic admittance of top Mn layer modelled as surface conductivity and characteristic input admittance of other two layers looking from top of Al_2O_3 (denoted as Y_2 in Fig. 2.2 (b)), (c) real and (d) imaginary part of permittivity of Mn along with those of the other 5 metals, extended up to 2000 nm wavelength.

So far, we have thoroughly investigated the effect of materials and highlighted the importance of the choice of metals. The above discussions show what a great potential Mn has to be employed in broadband absorption structures, even in higher wavelength ranges. Unlike its high potential, it has not been employed in absorbers so far. Therefore, we choose Mn for metal layers and Al_2O_3 as the dielectric layer for the structure, and proceed to the optimization of dimensions in the following section.

2.2.2 Dimension Optimization

As explained in the previous parts of this section, by increasing the thickness of the insulator layer, the optical path of the light inside the structure increases and the absorption band experiences a red-shift (see equation 2.1). One way to tailor the optical beam path ($n_i d_i$) is changing the dielectric material that indeed changes n_i in equation 2.1 that was explained in the previous section. The other method is the control of d_i .

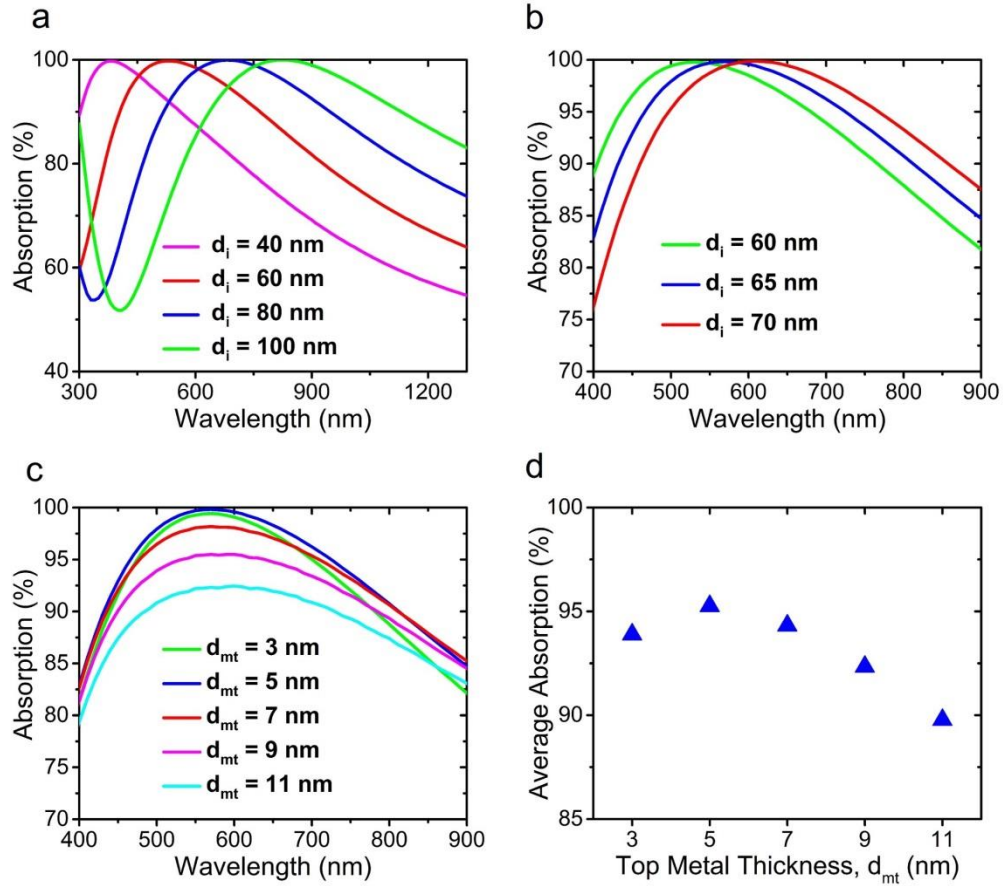


Figure 2.4: Calculated absorption spectrum of MIM structure for the different values of d_i with (a) 20 nm steps and, (b) 5 nm steps, while Mn is assumed as the metal and the top layer thickness is $d_{mt} = 5$ nm, (c) calculated absorption spectrum of MIM structure for different values of d_{mt} and, (d) average absorption for different values of d_{mt} calculated in the 400–900 nm wavelength range, again Mn is assumed as the metal and the thickness of the dielectric layer is $d_i = 65$ nm.

Fig. 2.4 (a) demonstrates the effect of the thickness of the insulator layer on the absorption spectrum. It can be observed that by choosing thicker dielectrics, the absorption band red-shifts. Among the calculated absorptions for different dielectric thick-

nesses, the one for 60 nm matches our desired spectrum, i.e. the visible range and some parts of NIR. For finely tuning the dielectric thickness, absorption spectrum has been calculated for different d_i values of 60, 65, and 70 nm as shown in Fig. 2.4 (b). As it can be seen from Fig. 2.4 (b), in the case of choosing 60 nm, the absorption strength will decrease in NIR range and, on the other hand, by choosing 70 nm as the dielectric thickness, absorption strength decreases in shorter wavelengths of visible range; but in the case of 65 nm, the best overall absorption strength and almost perfect absorption is obtained within the broad range of 400 nm to 900 nm wavelength. Therefore, d_i is chosen to be 65 nm.

Now, to choose the best thickness for the top metal layer or d_{mt} , absorption for different values of d_{mt} ranging from 3 to 11 nm has been calculated and is shown in Fig. 2.4 (c). It is obvious that the 5 nm is the best thickness for the top layer because it has the strongest and broadest response compared to the other d_{mt} values. Fig. 2.4 (d) illustrates the average absorption calculated in the 400 to 900 nm wavelength range versus d_{mt} , again demonstrating that 5 nm is the best choice for the top layer thickness.

To investigate the fabrication tolerance of the structure, which is a very important factor for mass production purposes, we have calculated the absorption of the structure for varying d_i versus wavelength and also for varying d_{mt} versus wavelength as shown in Fig. 2.5 (a) and (b), respectively. These color-plots show that deviations from optimal dimensions in the order of a few nanometers do not affect the absorption bandwidth and strength, significantly.

As for the bottom layer, there is no optimization needed and it is only necessary to be thick enough so that it almost completely blocks the transmission.

Fig. 2.5 (c) depicts the transmission for different thicknesses and varying wavelength and gives a good insight about this issue. There is a slight increase in transmission toward higher wavelengths that is due to the decrease of the optical path inside the material which leads to reduction of absorption of the light passing through it. Fig. 2.5 (d) shows the maximum and average transmission in the range of 400-900 nm versus Mn thickness. It is obvious that maximum transmission remains below 1 per-

cent for thicknesses down to around 100 nm. Based on these figures, we conservatively set the thickness of the bottom Mn layer to 200 nm.

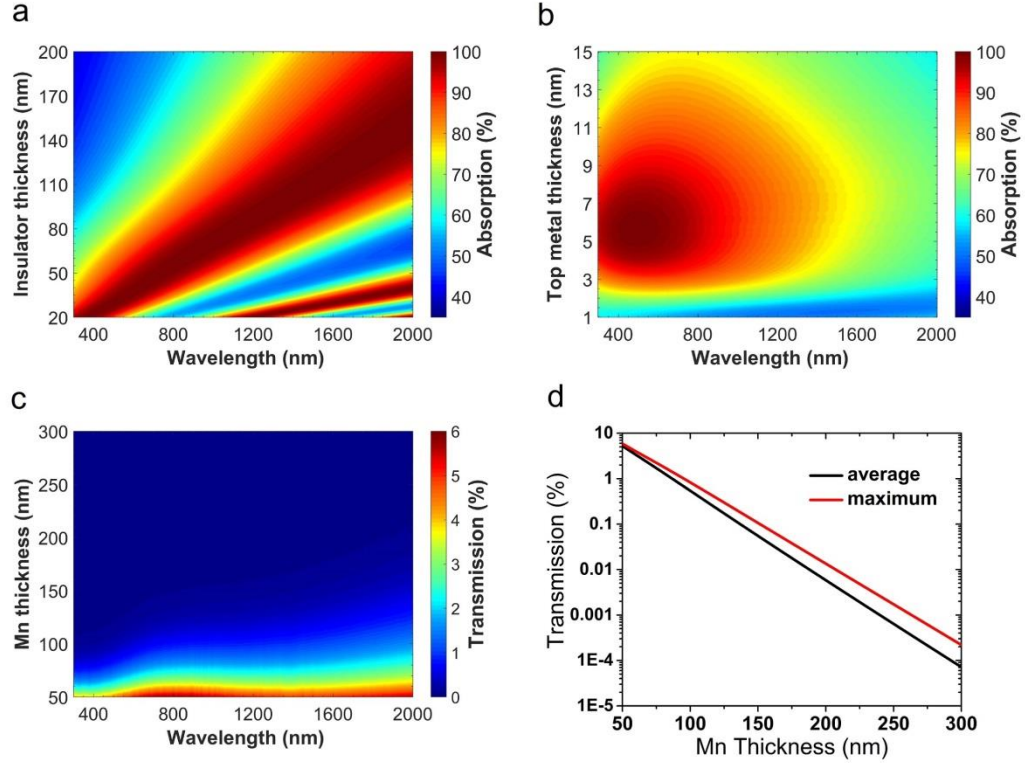


Figure 2.5: (a) Calculated absorption of MIM structure versus the wavelength and different values of d_i , assuming Mn as the metal with $d_{mt} = 5$ nm and Al_2O_3 as the dielectric, (b) same as (a) but for different values of d_{mt} and with $d_i = 65$ nm, (c) calculated transmission from a layer of Mn versus wavelength and different thicknesses of Mn layer and, (d) maximum and average values of transmission from a layer of Mn versus Mn thickness, calculated in the 400-900 nm wavelength range.

To summarize, the optimized structure is as follows: The dielectric is chosen to be Al_2O_3 and the metal is Mn. The dimensions of 5, 65 and 200 nm are chosen for the top metallic layer, dielectric insulator, and the bottom thick metal layer, respectively. In the next section, we present the experimental results for this structure.

2.3 Results and Discussion

The structure with optimized dimensions was fabricated and characterized. In order to avoid any contribution from the substrate to the absorption, and also to be able to measure the transmission of MIM, the structure has been fabricated on a transparent Sapphire substrate. Fig. 2.6 (a) shows the measured transmission, reflection and absorption of the structure for the case of normal incidence. The experimental result for absorption demonstrates over 94 percent average absorption in the broad range of 400-900 nm and has absorption as high as 99.6 percent at the wavelength of 626.4 nm. The image of the fabricated absorber is shown in the inset of Fig. 2.6 (a) which demonstrates the black color of the absorber due to the strong absorption of the visible light. Moreover, as expected, the measurements confirm that the structure has zero transmission.

The experimental results are compared against simulation results calculated with the circuit model, TMM and FDTD, in Fig. 2.6 (b). It is obvious that the simulation and experimental results are in good agreement. The insignificant mismatch between the simulation and experimental results may be due to using different deposition rates for the top and bottom metal layers (see Methods, Experimental). The permittivity profile of Mn has been extracted from a film deposited by the same rate as the bottom metal. Therefore, since using different deposition rates may affect the grain size which may consequently affect the optical properties, the optical properties of the top metal layer may be slightly different. However, this possible difference in the optical properties must have been insignificant because of the negligible mismatch between theory and experiment. Other possible causes may be variations in the thickness of the layers and also imperfections in the roughness of the top layer since it is very thin and it is hard to obtain ultra-thin metallic layers with no roughness while using physical vapor deposition methods. However, for broadband absorbers, roughness is a positive characteristic and contributes to the strength and also the broadening of the absorption because it leads to having the superposition of various modes in the absorption.

Now let us investigate the performance of the fabricated structure for oblique incidence and to show that it has an omnidirectional performance, which is very important in different applications. We measured the absorption of the structure for both TM and TE polarizations and for angles ranging from 20 to 70 degrees. Figures 2.6 (c) and 2.6 (d) show the absorption of the structure for the mentioned incident angles in the wavelength range of 400 to 900 nm, for TM and TE polarizations, respectively. It is obvious that the device retains its high and broadband absorption performance up to very large incident angles such as 70 degrees for TM polarization. However, the TE polarization is more sensitive to the variations of incident angle. Note that the range of y-axis is different for figures 2.6 (c) and 2.6 (d).

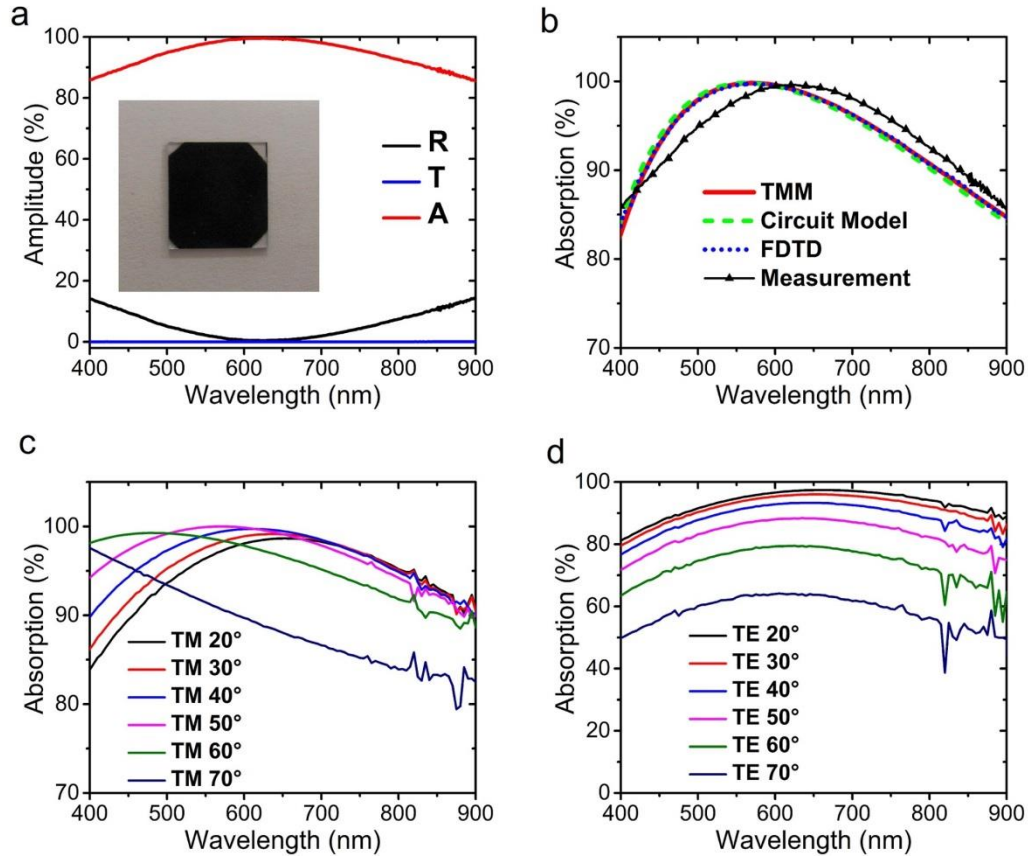


Figure 2.6: (a) Measured reflection, transmission and absorption spectrum of the fabricated MIM structure, inset: image of the fabricated sample, (b) comparison of simulation and calculation methods with measurement for the absorption spectrum of the optimal MIM structure, and, measured absorption of the MIM structure with different angles of incidence ranging from 20 to 70 degrees, for (c) TM and (d) TE polarizations of incident light. Note that y-axis range is different between (c) and (d).

To further elucidate the electromagnetic phenomena taking place in this structure, the electric field distribution inside the structure for the wavelength range of interest (400-900 nm) is depicted in Fig. 2.7 (a). Since the structure is not changing in the lateral direction, we have only depicted the vertical spatial variation of the field inside the structure. It is obvious that a cavity resonance mode is trapped inside the structure in the dielectric region and back and forth reflections from the top and bottom metallic layers lead to a standing wave inside it.

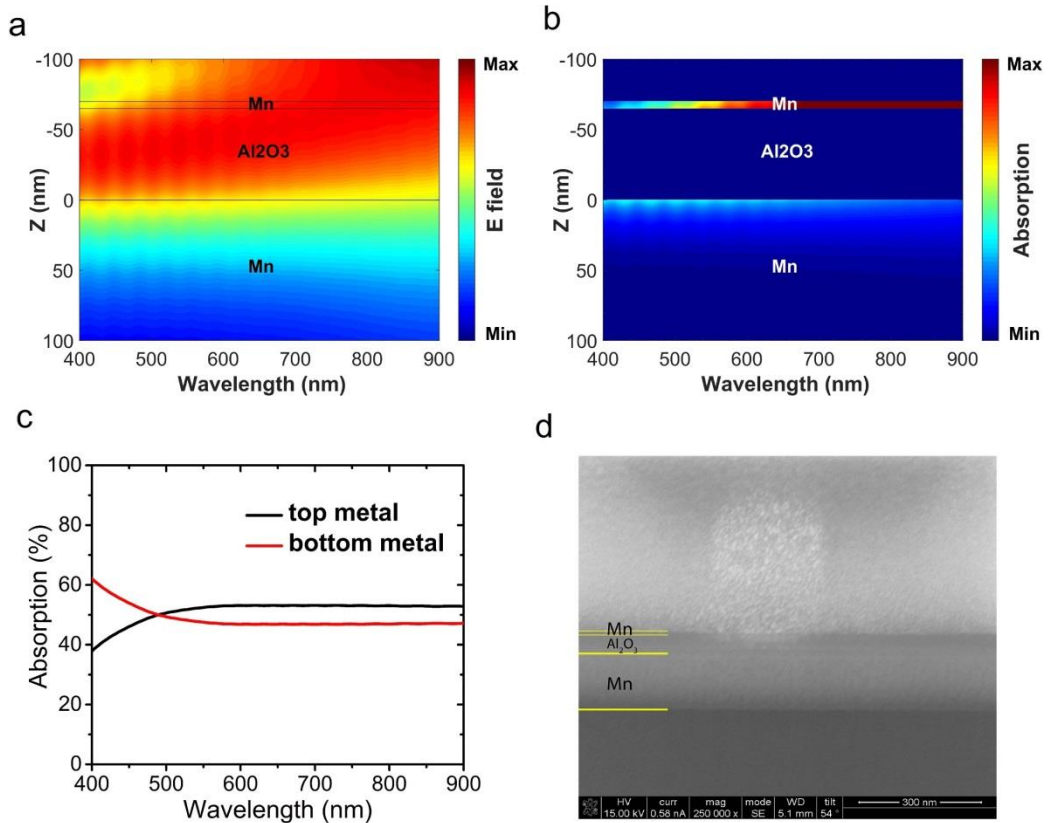


Figure 2.7: Simulated (a) electric field intensity and (b) absorption distribution in the cross section of the optimal MIM structure as a function of wavelength, (c) contribution of top and bottom metal layers in the light absorption and, (d) SEM image of the cross section of the fabricated MIM structure, milled by FIB

To better observe the contribution of each layer to absorption, the absorption intensity in different layers in the structure versus wavelength is shown in fig. 2.7 (b). It can be seen that the absorption intensity is high at the top metal, but it is accumulated in a thin layer. In the bottom metal, on the other hand, the absorption intensity is low-

er compared to the top metal, but it is distributed in a thicker layer. To evaluate the total contribution of each layer to absorption, Fig. 2.7 (c) shows the percentage of total absorption in each metallic layer, versus wavelength. It can be observed that the contribution of the top and bottom metallic layers is not significantly different. As can be seen in both figures 2.7 (b) and (c), at lower wavelengths, the bottom layer has a larger contribution to the absorption and the absorption of the top metal layer increases at longer wavelengths. Finally, the Scanning Electron Microscopy (SEM) image of the cross section of the fabricated sample, which is milled by Focused Ion Beam (FIB) is shown in Fig. 2.7 (d).

2.4 Conclusion

Step-by-step design and optimization of a broadband MIM absorber is presented, geometrically and in terms of materials. The important advantage of this absorber is the use of three large area layers without any need to lithography or any repetition of MI pairs, which makes this absorber as simple as possible. It has high absorption within the whole visible range and going into the NIR region. Achieving such a response with such a simple structure is very promising for mass production. We have used and introduced Mn for the first time as the metal layer in a broadband absorber. We have shown that Mn is a promising material for broadband absorption by investigating its permittivity profile. We have shown that its complex permittivity profile is very close to that of the ideal metal for achieving perfect impedance matching to that of free space in our wavelength range of interest (400-900 nm). It was shown experimentally that the structure has an omni-directional response up to angles as large as 70 degrees, for both TE and TM polarizations. This section proved that Mn has great potential in broadband absorption structures and their deriving applications.

2.5 Methods

2.5.1 Experimental

For the fabrication of the structure, first, a typical commercial square-shaped transparent Sapphire wafer with 100 mm^2 area has been cleaned with Piranha solution, de-ionized water, Acetone, and Isopropanol, and then has been dried with N_2 flow, to be used as the substrate. Then, a 200 nm Mn film has been coated on it using VAKSIS Thermal Evaporator in the chamber pressure of $3\text{e-}6$ to $5\text{e-}6$ Torr and with the rate of 0.5 A/s. After that, 65 nm Al_2O_3 has been coated using H_2O and $\text{Al}(\text{CH}_3)_3$ as precursors at 200°C in an ALD reactor (Cambridge Nanotech Savannah S100). Finally, 5 nm Mn has been coated on the structure as the top metal layer using the same Thermal Evaporator as for the bottom layer, at the same chamber pressure and with the rate of 0.1 A/s. For characterization, a home-made fiber optic based reflectometer setup has been used for measuring the reflection at normal incidence up to the wavelength of 850 nm. For wavelengths above 850 nm, measurement of the reflection at normal incidence has been carried out using Fourier Transform Infra-Red (FTIR, Bruker). Reflection measurements with angled incidence and for different polarizations, transmission measurements, and material characterizations for permittivity extraction have been done using J.A. Woollam Co. Inc. VASE Ellipsometer.

2.5.2 TMM Calculations

The calculations of the TMM method have been carried out by dividing the total electromagnetic wave inside each layer into sum of two waves each going in opposite directions, forward and backward, both of which are perpendicular to the interface of layers. Figure 2.1(a) depicts the 3D schematic of the structure and Fig. 2.1 (b) shows the cross section of the structure in xz plane. The structure is infinite in x and y directions and z axis is toward top to bottom, perpendicular to the interfaces of layers. The total electric field (E_i) and magnetic field (H_i) inside each layer can be written as:

$$E_i = E_i^+ e^{j\beta_i z} + E_i^- e^{-j\beta_i z} \quad (2.5a)$$

$$H_i = H_i^+ e^{j\beta_i z} + H_i^- e^{-j\beta_i z} \quad (2.5b)$$

where the subscript ‘i’ corresponds to the number of layer, which is a number from 0 to 4 (see Fig. 2.1 (b)), from top to bottom (including free space at both top and bottom sides of the structure with numbers of 0 and 4, respectively), E_i^+ and E_i^- correspond to the amplitude of the waves traveling forward (+z) and backward (-z), respectively. β_i is the complex propagation constant inside i^{th} layer which is equal to $\omega(\varepsilon_i \mu_i)^{1/2}$ with ω being the angular frequency, ε_i being the complex permittivity and μ_i being the complex permeability of the layer. μ_i is equal to permeability of free-space in the case of the materials we have used in this work (non-magnetic materials).

In order to relate the amplitudes in layer 0 (incident wave) to layer 4 (transmitted wave), we must multiply the interface and propagation matrices with the appropriate sequence. The propagation matrix of i^{th} layer, named as P_i , is the matrix which stands for the propagation of the wave inside i^{th} layer and transforms the field amplitudes at one end of the layer into another end, by multiplying them with their corresponding propagation coefficients, and is written as below for the i^{th} layer:

$$\begin{pmatrix} E_{(z_i)}^+ \\ E_{(z_i)}^- \end{pmatrix} = P_i \begin{pmatrix} E_{(z_{i-1})}^+ \\ E_{(z_{i-1})}^- \end{pmatrix} \quad (2.6)$$

$$P_i = \begin{pmatrix} e^{j\beta_i d_i} & 0 \\ 0 & e^{-j\beta_i d_i} \end{pmatrix} \quad (2.7)$$

where d_i is the thickness of i^{th} layer and z_i is the position on z axis at the end of the layer, as depicted in Fig. 2.1 (b).

To relate the field amplitudes in two sides of interface, boundary conditions should be imposed. Since the structure is symmetric in the xy plane, there is no difference between TM and TE waves in normal incidence and even one can assume the wave to be TEM. Without any loss of generality, We suppose the electric field to be in the +x

direction and, therefore, using the right hand rule, considering the fact that propagation is in the +z direction, the magnetic field has to be in the +y direction. Therefore, boundary conditions are indeed tangential boundary conditions that convey the continuity of electric and magnetic fields as follows:

$$(E_i^+ + E_i^-) \Big|_{z=z_i} = (E_{i+1}^+ + E_{i+1}^-) \Big|_{z=z_i} \quad (2.8a)$$

$$(H_i^+ + H_i^-) \Big|_{z=z_i} = (H_{i+1}^+ + H_{i+1}^-) \Big|_{z=z_i} \quad (2.8b)$$

By substituting H using the relation $H = (n/Z_0)E$ that holds between the electric and magnetic field, in which Z_0 is the impedance of free-space, and using the right hand rule, and after doing a few simplifications, equation (2.8b) evolves into the following equation:

$$(n_i E_i^+ - n_i E_i^-) \Big|_{z=z_i} = (n_{i+1} E_{i+1}^+ - n_{i+1} E_{i+1}^-) \Big|_{z=z_i} \quad (2.9)$$

Now, by having two equations of (2.8a) and (2.9) between electric fields in both sides of interface, we can write interface matrix named as $I_{i,i+1}$ which relates fields in two sides of the interface of i^{th} and $(i+1)^{\text{th}}$ layers, as follows:

$$I_{i,i+1} = \begin{pmatrix} \frac{n_{i+1} + n_i}{2n_{i+1}} & \frac{n_{i+1} - n_i}{2n_{i+1}} \\ \frac{n_{i+1} - n_i}{2n_{i+1}} & \frac{n_{i+1} + n_i}{2n_{i+1}} \end{pmatrix} \quad (2.10)$$

where

$$\begin{pmatrix} E_{i+1}^+ \\ E_{i+1}^- \end{pmatrix} \Big|_{z=z_i} = I_{i,i+1} \begin{pmatrix} E_i^+ \\ E_i^- \end{pmatrix} \Big|_{z=z_i} \quad (2.11)$$

Therefore, the relation between the field amplitudes of the 0th layer and 4th layer is in the form of

$$\begin{pmatrix} E_4^+ \\ E_4^- \end{pmatrix} = (I_{3,4})(P_3)(I_{2,3})(P_2)(I_{1,2})(P_1)(I_{0,1}) \begin{pmatrix} E_0^+ \\ E_0^- \end{pmatrix} \quad (2.12)$$

Then, we take E_0^+ equal to 1 as the amplitude of the incident wave, and set E_4^- to zero, since there is no backward propagating wave in free-space at the bottom of the structure, because of no reflecting interface after it. Therefore, there will be 2 knowns (E_0^+ and E_4^-) and two unknowns (E_0^- and E_4^+) that are related to each other by two equations generated from a 2 by 2 matrix.

Now, T and R of the MIM structure can be calculated as follows:

$$R = \left| \frac{E_0^-}{E_0^+} \right|^2 \quad T = \left| \frac{E_4^+}{E_0^+} \right|^2 \quad (2.13)$$

2.5.3 FDTD Simulations

FDTD simulations are carried out by employing Lumerical FDTD Solutions which is a commercial FDTD software package [113]. The simulations are done in a 2D simulation environment in the xy plane and a unit cell with 200 nm length in the x-direction is employed to define the structure. Boundary conditions are set to Periodic in the x axis boundaries and y axis boundaries are defined as Perfectly Matched Layer (PML). The structure is excited by a plane wave in the -y direction and a linear power monitor behind the source records the data of R. There is another linear power monitor on the other side of the structure that records T.

Chapter 3

Random Nanoholes for Broadband Absorption

3.1 Introduction

In this section, through annealing the MIM structure, we experimentally obtain over 94% average absorption in the ultra-wide range of 450-1400 nm that is a very promising result for a simple tri-layer lithography-free absorber. It is shown that annealing the sample leads to more than a twofold enhancement in the absorption bandwidth. We show that this is due to the fact that, after annealing, the top layer deforms into a metal-air composite with randomly sized, distributed and shaped nanoholes that support different Localized Surface Plasmon (LSP) resonance modes. The collective effect of the resonances of all random nanoholes leads to a significant absorption enhancement. The other feature contributing to such a broad absorption by such a simple structure is the optical properties of Mn that makes it an excellent candidate for broadband absorbers [114]. We experimentally demonstrate the absorption results for both cases of before and after annealing, and for a variety of incidence angles at both polarizations. This structure can serve as a very good candidate for large-area production of cost-effective absorbers covering a wide range of applications.

3.2 Design and Theory

Let us start with investigating the physical mechanism of the MIM resonant absorbers. Figure 3.1(a) demonstrates the schematic of the MIM configuration. The thickness of the layers from top to bottom is denoted as d_t , d_i , and d_b . The top layer must be optically thin enough to allow an appropriate portion of the incident beam penetrate into the cavity, and eventually get coupled to the cavity resonance mode. The resonant wave of the cavity for which the Fabry-Perot resonance condition is satisfied, will bounce back and forth between the top and bottom metallic layers. In each reflection a portion of the resonating wave is absorbed, which eventually leads to the perfect absorption of the light trapped inside the cavity. The Fabry-Perot resonance condition is as follows [115]:

$$2\left(\frac{2\pi}{\lambda_{res}}\right)n_i d_i + \varphi_b + \varphi_t = 2\pi m \quad (3.1)$$

where λ_{res} , n_i , φ_b , φ_t , m denote the resonance wavelength, refractive index of the dielectric material, phase-shift due to the reflection from the bottom metal, phase-shift due to the reflection from the top metal, and an integer number corresponding to the resonance order, respectively. The term $n_i d_i$ is the optical beam path traveled by the beam trapped inside the cavity, in each half-round of a full round-trip. Moreover, in our earlier work [15], it is shown in-detail how to calculate the φ_b and φ_t parameters. It can be observed from the equation that $n_i d_i$ (or the dielectric layer) mainly determines the spectral location of the absorption band, i.e. λ_{res} . It is noteworthy that typically, dielectric oxides such as SiO_2 , Al_2O_3 , HfO_2 , and TiO_2 have an almost constant refractive index in our desired wavelength range (visible and NIR). This means that for fixed metal layers, one can almost freely choose the dielectric material which determines n_i , and accordingly adjust the value of d_i to obtain the desired beam path inside the cavity, and consequently determine the absorption band location.

Since we can freely choose the dielectric oxide as explained above, we have chosen to use Al_2O_3 . The main reason for this choice is that this oxide can be coated using the atomic layer deposition (ALD) method. ALD devices perform atomic layer-by-layer depositions that in comparison with the physical evaporation-based deposi-

tion methods, result in much more conformal thin films. Moreover, by using this method, we have control over each atomic layer of the depositing material which provides us with a very high precision in obtaining the desired thickness [116]. It is noteworthy that the material and the thickness of the bottom and top metal layers also determine the values of φ_b and φ_t , respectively, thus affecting λ_{res} .

In the previous section, it was experimentally demonstrated that Mn has a superior performance in the MIM broadband absorbers compared to the other appropriate candidates for broadband absorption including Cr, Pt, Ti, and W [114]. It was explained that this is due to the fact that Mn has a small real part of permittivity which leads to high field penetration and has a large imaginary part of permittivity which is the cause of strong absorption. Therefore, Mn is chosen for both metal layers in our MIM configuration. In addition, unlike expensive metals such as Platinum, Gold, or Silver, Mn is an economically cost-effective and environmentally-friendly metal which has the potential to be used for the large-scale and mass production of this device [117].

Both the circuit model and FDTD simulations have confirmed that, to have strong absorption in the desired wavelength range that starts from visible and exceeds to NIR, for the case of using Mn and Al_2O_3 , the optimal dimensions are as $d_t = 5$ nm and $d_i = 70$ nm. However, in the previous section, it has been shown in detail that the absorption of the broadband MIM structure has a good fabrication tolerance for relatively small changes in the value of d_i [114]. The bottom layer only needs to be sufficiently thick enough to block any transmission so that we have conservatively chosen $d_b = 200$ nm. Absorption (A) in the general case is calculated from $A=1-R-T$ where R and T denote the reflection and transmission, respectively. In our case, T is zero so the absorption can be simply calculated from $A=1-R$.

3.3 Results and Discussion

The measured and calculated absorption spectrum of the absorber before annealing, at normal incidence is shown in Fig. 3.1(b). The calculations are carried out using the circuit model, the details of which can be found in [114]. The refractive index values

of Al_2O_3 and Mn used in the calculations are obtained by fitting the spectroscopic data from ellipsometry measurements. It can be observed that the calculations are in good agreement with the experimental results. The structure experimentally gives above 94% average absorption in the broad range of 450-900 nm. The scanning electron microscope (SEM) image of the cross-section of the structure milled by focusing ion beam (FIB) is shown in the inset of Fig. 3.1(b). Measured absorptions for TM and TE polarizations with oblique incidence are also shown in Figs. 3.1(c,d), respectively.

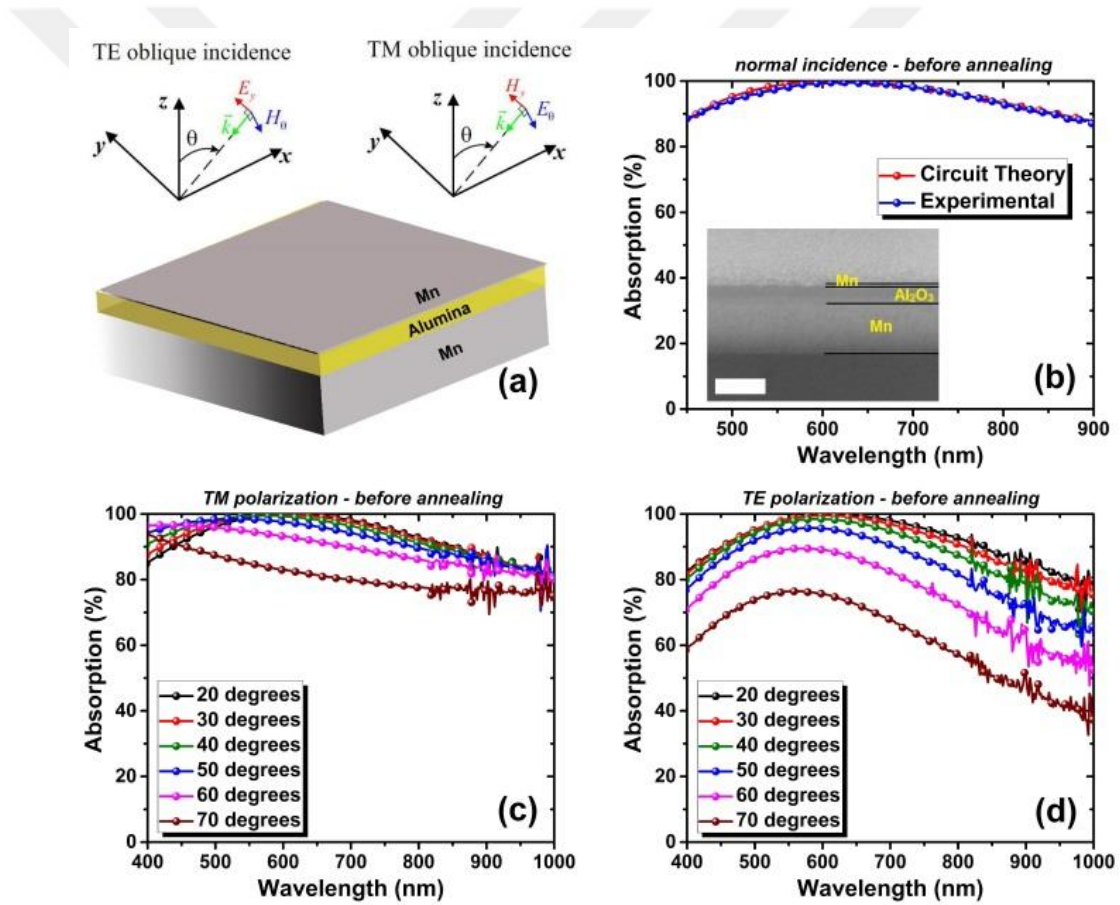


Figure 3.1: (a) Schematic of the MIM absorber, (b) Measured and circuit theory-based calculated absorption of the structure for normal incidence, inset: SEM image of the cross-section of the fabricated MIM absorber, milled by FIB; the white scale bar represents 200 nm. The measured absorption spectra with different angles of incidence for (c) TM and (d) TE polarization. All of the figures correspond to the MIM sample before annealing.

In order to obtain randomly sized and distributed nanoholes in the top metal layer that can support LSP resonances which lead to the absorption enhancement, we can use the dewetting method [118, 119]. Dewetting is a lithography-free process in which the ultra-thin metallic layers get annealed at high temperatures to obtain nano-patterns (nanoholes or nanoislands, based on the annealing recipe) in them. The randomness of the size, shape, and distribution of the nanoholes leads to the existence of various LSP modes with different spectral positions, the superposition of which, contributes to the broadening of the absorption band.

To obtain nanoholes in the top thin Mn layer, we performed the dewetting process on our fabricated MIM samples by examining different annealing recipes. The best result, which is presented in this letter, belongs to the case of annealing the sample for 5 minutes at a temperature of 500°C. The rise time to reach that temperature was fixed to 5 minutes and the temperature falling time (sample cooling period) was set to 1 minute. It is noteworthy that annealing at lower temperatures resulted in an insignificant absorption enhancement, and higher temperatures led to losing the absorption strength compared to the best annealing recipe. The SEM images of the top view of the MIM sample before and after annealing are shown in Fig. 3.2(a). It can be clearly observed that there is a high randomness in the size, shape, and distribution of the nanoholes. The measured absorption spectrum of the annealed sample for the case of normal incidence is shown in Fig. 3.2(b). For the purpose of a clear visualization of the absorption band broadening, the measured absorption spectrum of the sample before annealing is also included in this figure. It can be seen that the absorption band has experienced a significant enhancement compared to before annealing. The absorption has above 94% average value in the broad wavelength range spanning from 450 to 1400 nm. Therefore, compared to before annealing, the upper limit of the absorption band of above 94% average absorption, is increased from 900 to 1400 nm, while the lower wavelength limit is fixed at 450 nm, i.e. a 500 nm or over twofold bandwidth enhancement is experimentally achieved. The measured average absorption in the same range of 450-1400 nm was 83% before annealing. Such an achievement is obtained by only adding a simple and high-throughput annealing stage to the fabrication process. The normal incidence reflection measurements are carried out using a reflectometer and fiber optical cable-based home-made setup for up to 900 nm wavelength, and for larger wavelengths, measurements are done by a Fourier

transform infrared (FTIR, Bruker) device. Inset of Fig. 3.2(b) shows the image of the annealed fabricated sample which appears with black color due to the strong light absorption.

Figures 3.2 (c,d) demonstrate the measured absorption for oblique incidence when the incident wave is TM and TE polarized, respectively. It can be observed that the absorption band for oblique incidence has also experienced a significant enhancement

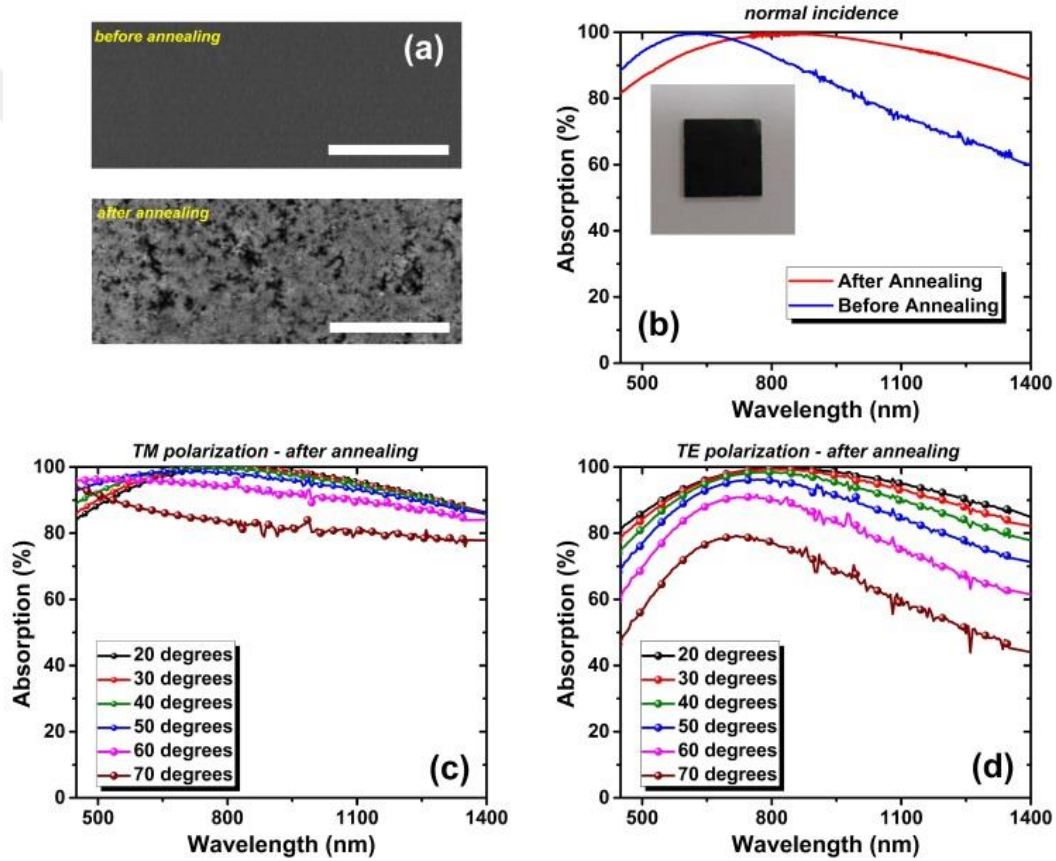


Figure 3.2: (a) SEM images taken from the surface of the MIM sample, before and after annealing; the white scale bar represents 2 microns. (b) measured absorption of the sample before (red line) and after (blue line) annealing for the case of normal incidence, and, for different angles of incidence in the cases of (c) TM and (d) TE polarizations. Inset of (b) shows the image of the annealed fabricated sample.

by annealing. The results show that, for the TM polarization, the absorption remains high for large angles up to 70 degrees, while for the TE polarization, the structure almost loses its high absorption performance after 50 degrees. In other words, the overall behavior of the structure for oblique incidence indicates that the structure has very low incidence angle sensitivity, especially for the TM polarization.

To observe the physical mechanism behind this broadband absorption, the SEM image of the annealed structure is imported into the Lumerical FDTD solver software [113] and the simulation is performed by defining it as the top layer. A square area of $1 \mu\text{m}^2$ from the SEM image is chosen for the simulation, and the periodic boundary conditions are defined in the both x and y directions. It is noteworthy that the new value for the thickness of the top layer can be approximated as $d_{ann} = (d_t/f)$, where $f = 0.8$ is the filling factor of the metal in the top layer's metal-air composite that is calculated from the taken SEM images. Therefore, since $d_t = 0.8$, the value of d_{ann} is calculated and considered as 6.25 nm in the FDTD simulations of the annealed structure. This is due to the fact that because of the appearance of the nanoholes, we can assume with some approximations that the top layer's thickness has increased in average. Figures 3.3(a,b,c) demonstrate the magnitude of the magnetic field, electric field, and absorption intensity, respectively. The field monitor is placed in the xy-plane cross-section at center of the top layer, with regards to the z-axis, and simulation results are shown for the four different wavelengths of 600, 800, 1000, and 1200 nm.

Magnetic field patterns shown in Fig. 3.3(a) provide a clear explanation for the role of random nanoholes in absorption band enhancement. As can be seen, the field gets localized, enhanced, and confined inside the nanoholes, which is demonstrative of LSP mode excitations. It can be observed that for the cases of different wavelengths, different nanoholes with different sizes get excited. To explain this phenomenon, relatively large and observable nanoholes are numbered from 1 to 5 in Fig. 3.3(a). It can be seen that for instance, at the wavelength of 600 nm, mainly nanoholes 2, 4, and 5 get excited and field localization at the other nanoholes is not as strong as these ones. On the other hand, for the wavelength of 1000 nm, mainly nanoholes 1 and 3 get excited. Therefore, as mentioned above, at any chosen wavelength in the absorption band, specific nanoholes with specific sizes get excited, and as the wavelength increases, it is observable that larger sized nanoholes get excited, which is in total agreement with the nature of LSP resonance modes [120]. Consequently, the superposition and collective effects of all randomly-sized nanoholes existing in the top metallic layer lead to a significant absorption band widening in a broad wavelength range.

It can be observed from the electric field magnitude pattern in Fig. 3.3(b) that there is a field localization and enhancement in the nanohole edges for all wavelengths. As will be clearly explained later, such a field localization at the nanoholes edges which include metal-air interfaces, leads to an absorption enhancement at these points.

The absorption intensity patterns shown in Fig. 3.3(c) are calculated using the following formula:

$$P_{abs} = -0.5\omega|E|^2 \text{imag}(\epsilon) \quad (3.2)$$

where ω is the angular frequency, $|E|$ is the electric field magnitude, and ϵ is the permittivity of the material. It is obvious that the absorption will be zero for the air regions inside the nanoholes, and for a constant wavelength, i.e. constant ω , the absorption intensity in the metallic parts with the same metal will only be a function of $|E|^2$.

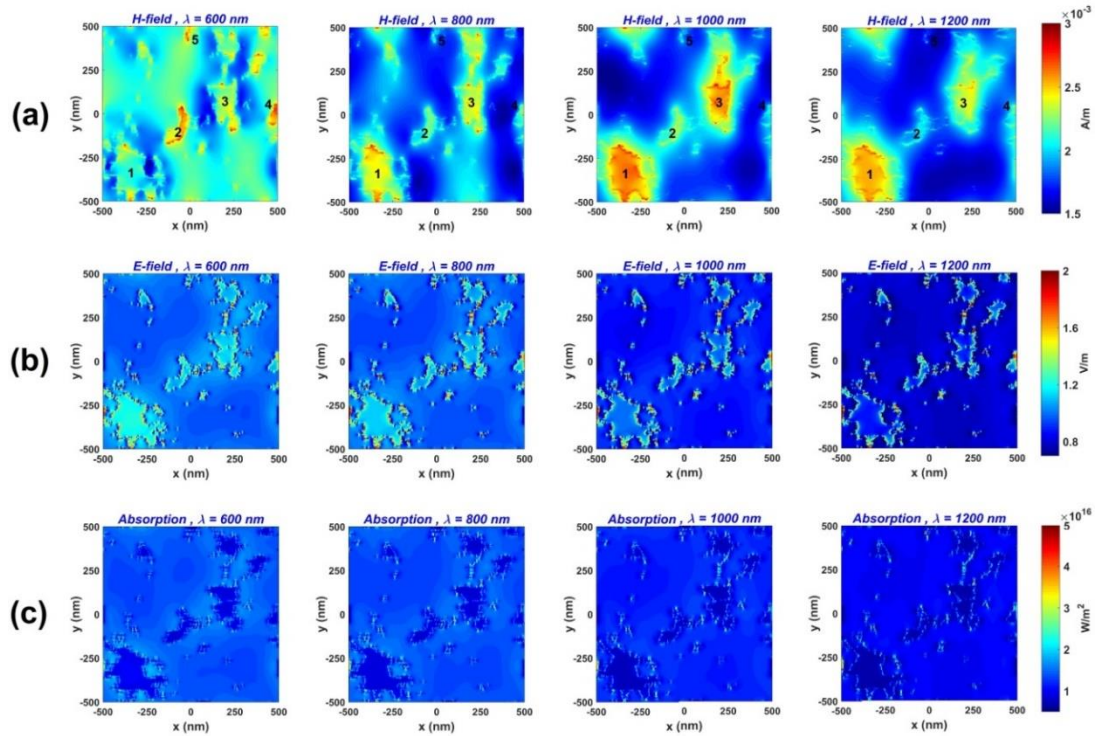


Figure 3.3: (a) Magnetic field magnitude, (b) electric field magnitude, and (c) absorption intensity distribution of on the xy cross-section of the center of the top metal layer of the annealed structure, simulated by importing the SEM image of the sample into the Lumerical FDTD solver.

This is the reason why the absorption intensity patterns of the metallic parts in Fig. 3.3(c) are relatively similar to the electric field magnitude patterns in Fig. 3.3(b), except with the explained differences. It can be clearly observed that a significant absorption enhancement happens at the hotspots of the edges of the nanoholes on the top layer which is the result of field localizations due to the excitation of LSPs, shown in Fig. 3.3(a).

3.4 Conclusion

In summary, an ultra-broadband tri-layer MIM absorber is fabricated and characterized. We have shown that, through annealing, an above 94% average absorption is experimentally achieved in the wide range of 450-1400 nm, which is a very promising result compared to the simplicity of the structure. It is explained that this is due to the deformation of the top layer into metal-air composite that includes random nanoholes with the potential of supporting different of LSP resonance. Moreover, it is shown that at different wavelengths, different nanoholes get excited and a collective effect of all nanoholes contributes to such a significant absorption enhancement. It is experimentally demonstrated that this absorber has very low angle sensitivity for the TM polarization up to 70 degrees, and for the TE polarization up to 50 degrees. The structure is lithography-free and cost-effective, so it is a perfect candidate for large-area production and utilization in photovoltaic and solar cell applications.

Chapter 4

Patterned MIM Embedded with Ultra-Thin Manganese Film

This chapter is on part reprinted with permission from: **Majid Aalizadeh**, A. Khavasi, A. E. Serebryannikov, G. A. E. Vandenbosch, and E. Ozbay, “A Route to Unusually Broadband Plasmonic Absorption Spanning from Visible to Mid-infrared,” *Plasmonics*, 2019.

4.1 Introduction

One of the commonly used absorber structures is the MIM structure, in which the top metal layer is patterned using lithography. In one of the recent works, Au-SiO₂-Ti configuration is used in the MIM structure, and the top Ti layer is patterned as a periodic array of nanodisks [36]. The high absorption band in that structure is extended over the wavelength range of 900 to 1,825 nm with a 71% percent relative bandwidth that is defined as the ratio of the high absorption bandwidth to the center wavelength.

In this section, we show that, by embedding an ultra-thin 5 nm layer of Mn into the dielectric layer of a typical top-layer-patterned MIM structure (with a 2D array of nanodisks on the top), an ultra-broadband, nearly perfect absorption (absorption greater than 0.9) can be obtained over the wavelength range of 478 to 3,278 nm. The bandwidth and relative bandwidth of the proposed structure are 2,800 nm and 149%,

respectively, and the absorption band extends over the visible, NIR, and MIR regions. This is, to our knowledge, the best result obtained for such absorbers with one-stage-lithography. The absorption band of the MIM structure without the ultra-thin Mn layer covers the two adjacent ranges of 478 to 526 and 610 to 836 nm that is equivalent to 274 nm of total bandwidth. Therefore, adding a thin layer of Mn, leads to a 10.1 fold increase in the bandwidth, without adding much complexity or lithography stage to the fabrication process. A step-by-step dimension and material optimization procedure is presented. We investigate different types of metals in order to find the most appropriate ones for an ultra-wideband absorber. We also consider the effect of the dimensions of the structure on the absorption bandwidth. Then, the physical mechanisms that lead to strong absorption in different wavelength ranges are discussed. In particular, we discuss the importance of the 5 nm Mn layer for the significant increase of the bandwidth of the structure. This is while Mn has not been given the attention it deserves for the use in broadband absorbers. Mn was used for the first and, up to now, the only time in a broadband absorber structure that was recently fabricated in our group, and it led to a very promising result compared to other metals [114]. Moreover, we demonstrate the high fabrication tolerance of the proposed structure. Finally, we explore the effect of polarization and incidence angle. Numerical results are obtained using the finite difference time domain (FDTD) method by Lumerical FDTD software [113], and the results are verified by regenerating them using the finite integration technique (FIT) by CST Microwave Studio [121].

It is worth mentioning that, because of requiring just one-step lithography, the large-area fabrication of the proposed structure can be realized by the use of the standard nanoimprint lithography technique [122]. This is very beneficial for mass and high-throughput production purposes.

4.2 Design and Theory

4.2.1 Proposed Structure

The schematic of the proposed absorber is shown in Fig. 4.1. It consists of an MIM structure with the top and bottom metal layers being chosen as Ti, and Al_2O_3 being used as the dielectric layer, and with the top Ti layer being patterned to obtain a periodic array of nanodisks, while an ultra-thin, 5 nm layer of Mn is embedded into the dielectric layer. As shown in Figs. 4.1(a-c), the period, height of nanodisks, radius of nanodisks, thickness of the Mn layer, the distance between the bottom metal and the top nanodisks, and the distance between the bottom metal and the Mn layer, are denoted by p , h , r , t_{Mn} , d_i and d_{Mn} , respectively. The bottom metal layer is expected to be sufficiently thick to block transmission. The perspective description of the proposed absorber is shown in Fig. 4.1(d). In this view, the 5 nm Mn layer can be considered as a perturbation that is introduced to the patterned MIM configuration of Ti- Al_2O_3 -Ti. This is while, as it will be elevated in the forthcoming sections, the effect of this ultra-thin Mn layer is drastically significant in the enhancement and broadening of the nearly perfect absorption band. The optimized dimensions for the structure, according to Fig. 4.1, will be obtained in the next sections. They are equal to 480, 140, 180, 165, 5, and 135 nm for p , r , h , d_i , t_{Mn} , and d_{Mn} , respectively. The thickness of the bottom metal layer is conservatively chosen to be 200 nm to ensure nearly zero transmission. It should be mentioned that this choice of metals is the result of simulating all the possible combinations of several metals (which are conventionally used in broadband absorbers) in the proposed structure, and performing a thorough study on the results. These metals include Chromium (Cr), Tungsten (W), Platinum (Pt), and Iron (Fe), in addition to Ti and Mn.

4.2.2 Material Optimizations

In order to demonstrate the effect of the choice of materials on the performance of the device and optimize the structure in terms of materials, the absorption spectra have been simulated. The results are shown in Figs. 4.2(a), (b), (c), and (d) for the case of

only changing the material of the dielectric layer, bottom metal layer, top nanodisks, and the 5 nm ultra-thin middle layer, respectively. All of the other materials and dimensions of the structure were kept unchanged. The thin horizontal dashed line in Fig. 4.2 corresponds to 90 percent absorption. The refractive index data of all the metals except Mn, along with Al_2O_3 and SiO_2 are taken from [123], and the refractive index data of Mn, HfO_2 , and TiO_2 are taken from [124], [125], and [126], respectively. Our purpose here is to choose the configuration that gives the widest band, while the bandwidth (BW) is defined as $\text{BW} = \lambda_{\text{max}} - \lambda_{\text{min}}$, where λ_{max} and λ_{min} are the upper and lower wavelength limits of the region in which absorption is above 90%. The absorption (A) in general case can be obtained given the values of reflection (R) and transmission (T), using the formula $A=1-R-T$, where $R=|r|^2$ and $T=|t|^2$ with r and t being reflection and transmission coefficients, respectively. However, since the bottom metallic layer is sufficiently thick to suppress any transmission (200 nm), the absorption formula simplifies to $A=1-R$.

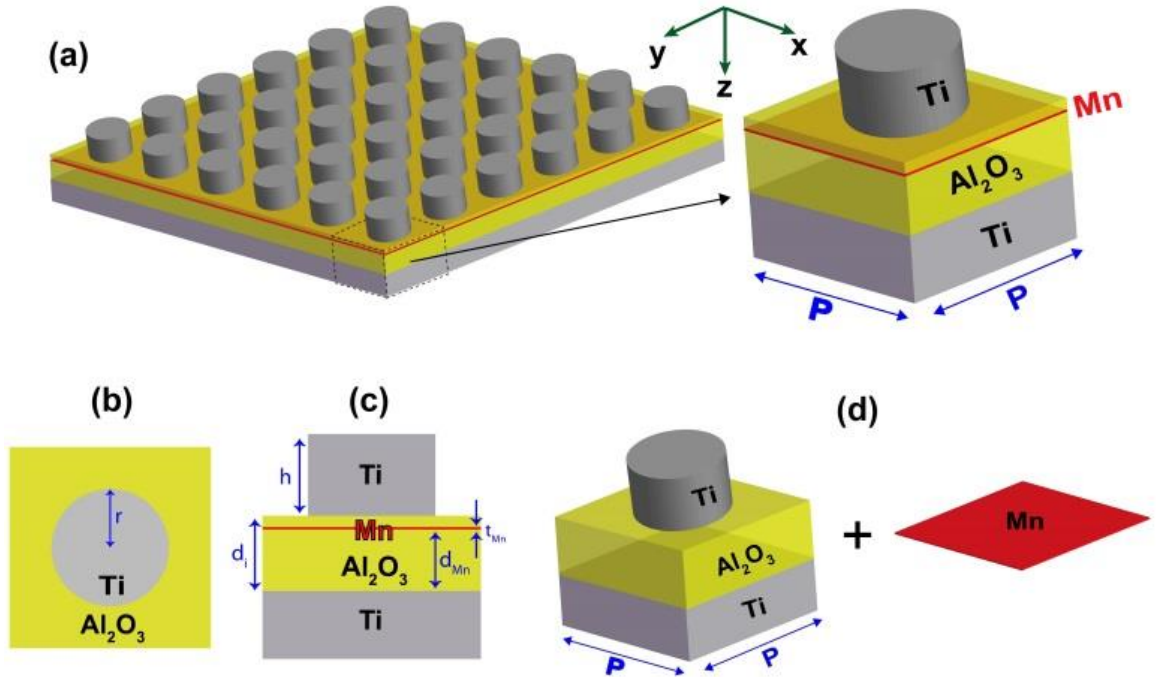


Figure 4.1: (a) Left plot: 3D schematic of the absorber; one unit cell of the structure is bounded by dashed lines; right plot: a magnified view of one unit cell. (b) Top view, and (c) side view of the structure; and (d) the other image of the unit cell of the studied structure (perspective view): a patterned MIM configuration with top periodic

layer composed of metal nanodisks, plus an ultra-thin Mn layer embedded into the insulator of the original configuration.

Let us first consider the effect of dielectric material on the absorption spectrum. The dielectrics chosen to be used in this investigation are SiO_2 , Al_2O_3 , HfO_2 , and TiO_2 , which are listed from smaller to larger refractive indices, respectively. It can be observed in Fig. 4.2(a) that by choosing dielectrics with higher refractive index, almost all the absorption peaks experience a red-shift. This is already expected to happen because of the nature of the resonance modes [20]. This figure demonstrates that the best bandwidth is obtained for the case of using Al_2O_3 as the dielectric material. For the case of changing the materials of the bottom metal and the top nanodisks, as shown in Figs. 4.2(b) and (c), respectively, it can be noticed that the absorption does not experience drastic changes. This shows that the absorption capability of the entire structure, when taking the bottom layer and top nanodisks into account, dominantly results from the geometric design rather than the choice of materials. However, about the middle metal layer, which is taken as 5 nm, it can be observed in Fig. 4.2(d) that the choice of metal drastically affects the absorption spectrum's shape and bandwidth at wavelengths larger than 800 nm. Therefore, this ultra-thin layer plays an important role in broadening the absorption spectrum in the NIR and MIR regions. The results shown in Figs. 4.2(a-d) justify that the choice of the materials in Fig. 4.1 provides the widest absorption band.

4.2.3 Dimension Optimizations

To demonstrate the effect of the radius of the nanodisks (r) and the period (p), the absorption spectrum is calculated and shown for different values of r , for $p = 440, 460, 480, 500, 520$ and 540 nm, in Figs. 4.3(a), (b), (c), (d), (e), and (f), respectively. It is noteworthy that h is fixed at 180 nm, and the materials are chosen in accordance to Fig. 4.1. Similarly, calculations are performed for the case of varying the height of nanodisks (h), and for above-mentioned values of p at $r = 140$ nm. The results are shown in Figs. 4.4(a-f). As shown in Fig. 4.3, by increasing the radius of nanodisks, the second, third, and fourth absorption maxima are shifted toward larger wavelengths. However, it comes at the cost of losing the strength of absorption at smaller

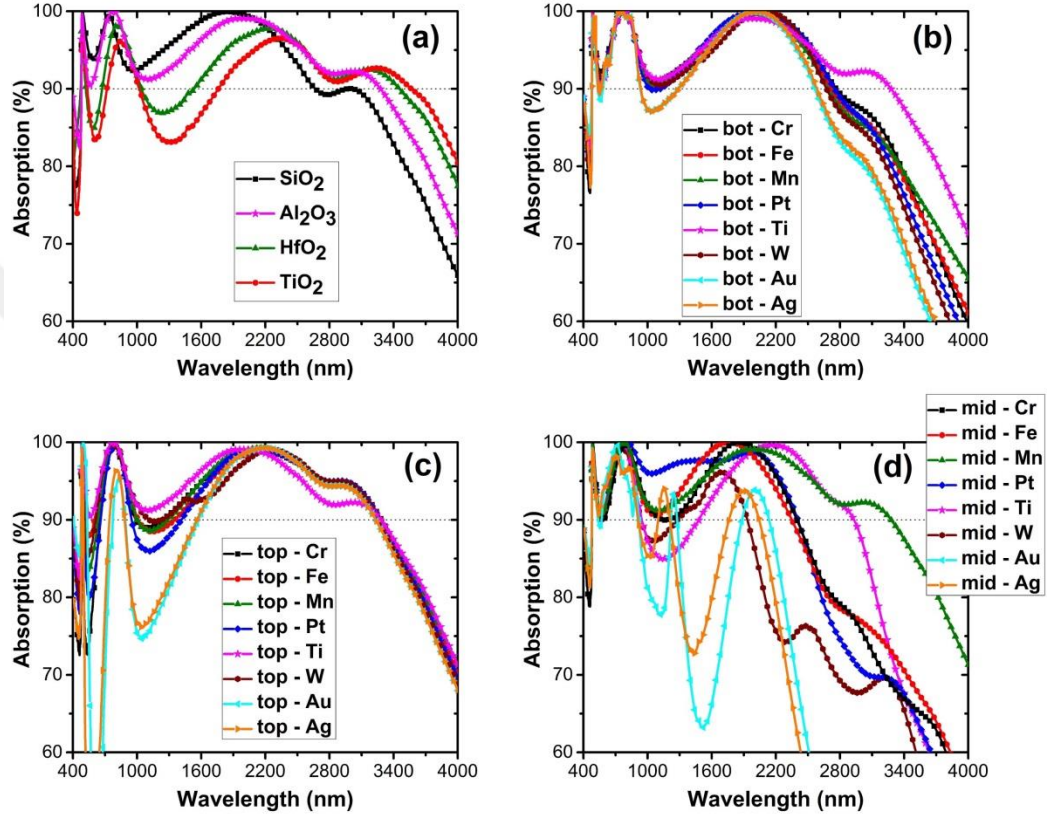


Figure 4.2: Absorption spectra for different materials of the (a) dielectric layer, (b) bottom metal layer, (c) top metallic nanodisks, and (d) middle ultra-thin metal layer, while using the same materials for the remaining components as in Fig. 4.1. The dimensions of the structure in all cases are: $p = 480$ nm, $r = 140$ nm, $h = 180$ nm, $d_i = 165$ nm, $t_{Mn} = 5$ nm, and $d_{Mn} = 135$ nm. The geometrical sizes in case of another material of the middle layer than Mn in (d) are the same as in case of Mn.

wavelength range. In addition, a comparison of Figs. 4.3(a-f) reveals that increasing the period of the structure blue-shifts the absorption band and increases the strength of the absorption in small wavelength regime. By comparing the bandwidths for all the presented cases, we choose the case presented in Fig. 4.3(c) where $p = 480$ nm and $r = 140$ nm, which provides the largest absorption bandwidth. As can be seen in Fig. 4.4, the increase in the height of nanodisks exerts the same effect on the absorption spectrum as the increase of r . Again, the results show that the best values for h

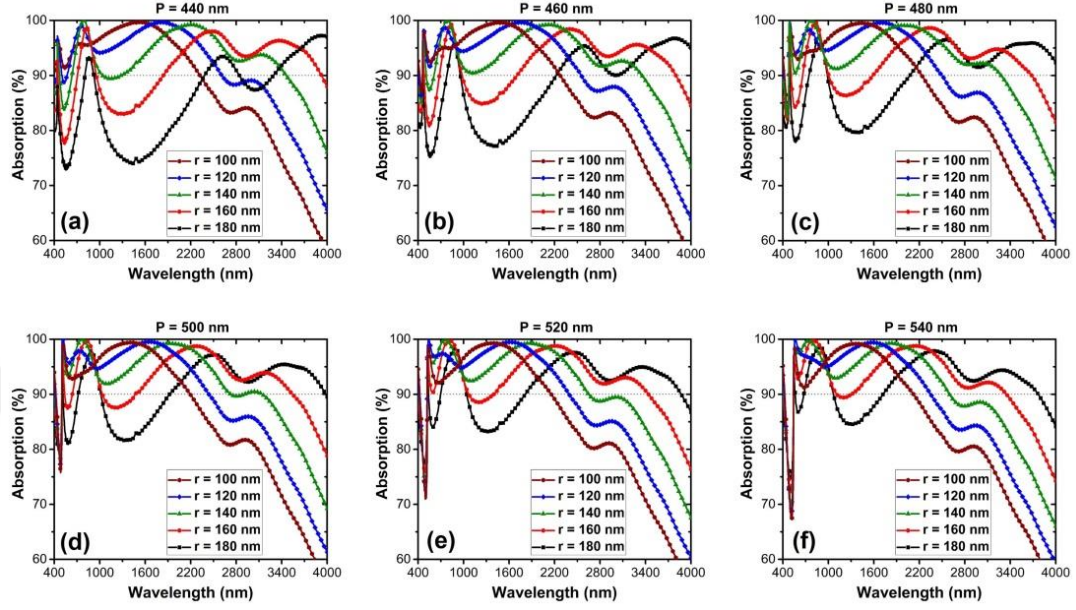


Figure 4.3: Absorption spectra of the structure at different values of the radius of nanodisks (r), while period (p) is taken as (a) 440, (b) 460, (c) 480, (d) 500, (e) 520, and (f) 540 nm. In all figures, $h = 180$ nm and the materials are chosen in accordance to Fig. 4.1.

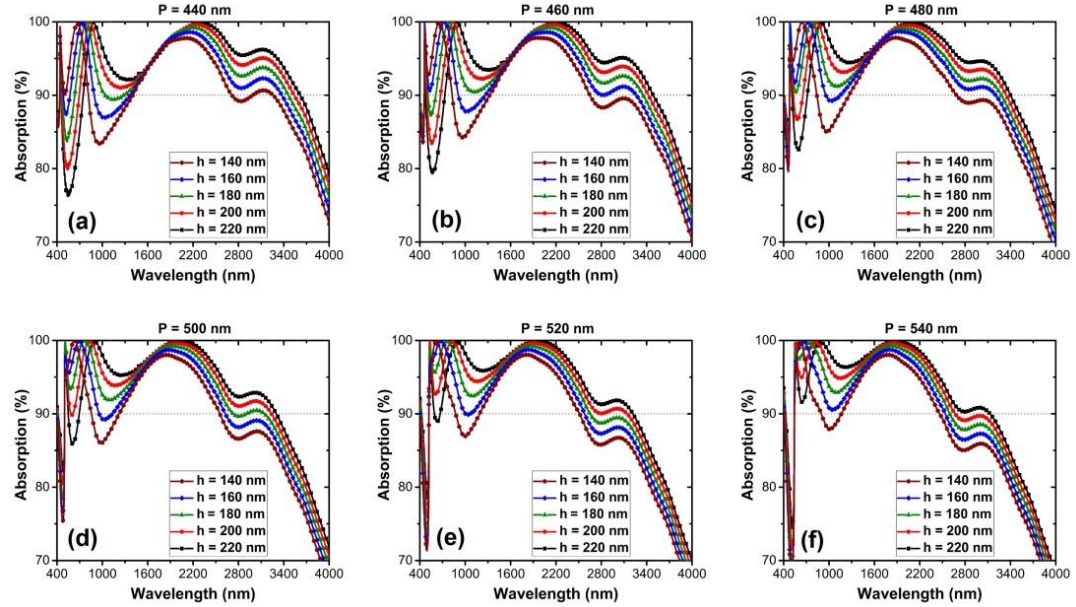


Figure 4.4: Absorption spectra of the structure at different values of the height of nanodisks (h), while period (p) is taken as (a) 440, (b) 460, (c) 480, (d) 500, (e) 520, and (f) 540 nm. In all figures, $r = 140$ nm and the materials are chosen in accordance to Fig. 4.1.

and p are 180 and 480 nm, respectively. Therefore, Figs. 4.3 and 4.4 confirm that the optimal values for r , h , and p are 140, 180, and 480 nm, respectively.

4.3 Results and Discussion

4.3.1 Absorption of the Optimal Design

The absorption spectrum of the optimal structure is plotted in Fig. 4.5. To verify the FDTD simulations, the absorption of the structure has also been calculated using CST Microwave Studio software, which is based on the FIT. The FDTD and FIT results are shown with solid red and dashed black lines, respectively. It is shown in Fig. 4.5(a) that the high absorption (absorption above 90%) region starts at the visible, covers the whole NIR, and ends at the MIR regime, i.e. it is in the wavelength range extending from 478 to 3,278 nm. The bandwidth is then 2,800 nm. The relative bandwidth of absorption is calculated by $RBW = BW/\lambda_{cent}$, where BW and λ_{cent} denote the bandwidth and the center wavelength of the absorption band, respectively. For this structure, we obtain $RBW = 149\%$. This is, to our knowledge, the best result obtained for such absorbers with one-stage lithography that may cover a very wide wavelength range, which starts at the visible and ends within the MIR range.

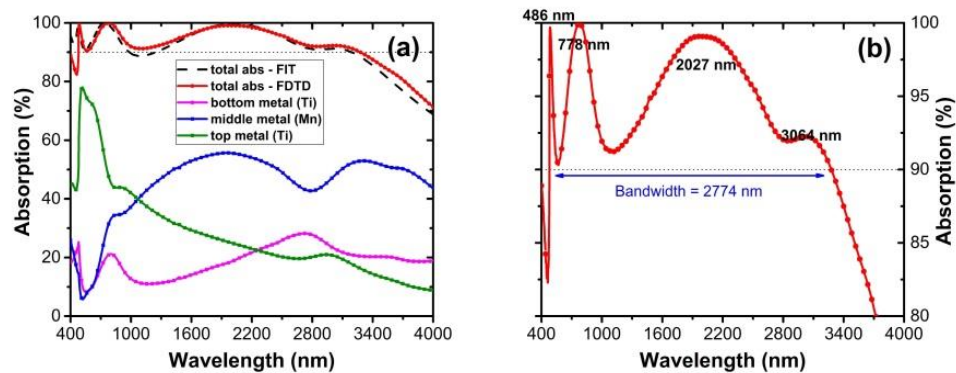


Figure 4.5: (a) Absorption spectra of the optimal structure calculated by FDTD (solid red line) and FIT (dashed black line) methods, along with contribution of each metallic layer to the absorption of the entire structure, and (b) absorption of the optimal structure (FDTD results) with magnified ordinate axis, to clearly demonstrate the peak positions and bandwidth of the absorption.

The contribution of each metallic layer (as an absorptive component) to the resulting absorption of the structure versus the wavelength is also plotted in Fig. 4.5(a), where the absorption spectra of the bottom Ti layer, ultra-thin Mn layer, and top Ti nanodisk array are shown as pink, blue, and green lines, respectively. The absorption of each individual layer of the entire structure is calculated by placing the field recording and index recording monitors inside the structure in the simulation, and calculating absorption at multiple observation points inside each layer.

4.3.2 Physical Analysis

In this section, we investigate the physical mechanism behind the observed wideband absorption. As shown in Fig. 4.5(b), the absorption peaks appear at 486, 778, 2,027, and 3,064 nm. The peak at the wavelength of 3,064 nm shows weaker absorption compared to the other peaks, but as it can be seen in Fig. 4.2(d), this peak helps the nearly perfect absorption bandwidth to increase significantly compared to the other possible choices of metals for the ultra-thin layer. Therefore, it is worth discussing the physical mechanism behind this peak, as well as the other peaks.

As the first step, we consider the absorption of each metallic layer versus the wavelength as shown in Fig. 4.5(a). It can be observed that at smaller wavelengths, up to around 1,000 nm, the absorption in the Ti nanodisks dominates the absorption of the Mn layer and the bottom Ti layer. This conveys that the absorption at smaller wavelengths must be mainly due to some resonance modes occurring close to the nanodisks. These resonances may lead to the absorption of light around the nanodisks, which are made of Ti, a highly lossy metal that provides the main contribution to the resulting absorption. On the other hand, at larger wavelengths, i.e. nearly after 1,000 nm, a great portion of light is absorbed by the ultra-thin, 5 nm layer of Mn, while the absorption portions of the Ti nanodisks and the thick bottom Ti layer are relatively smaller. This occurs despite the fact that the both Ti components have a much larger thickness than the Mn layer. This again signifies the crucial role of the Mn layer in the broadness of the absorption spectrum that is achieved in the studied structure.

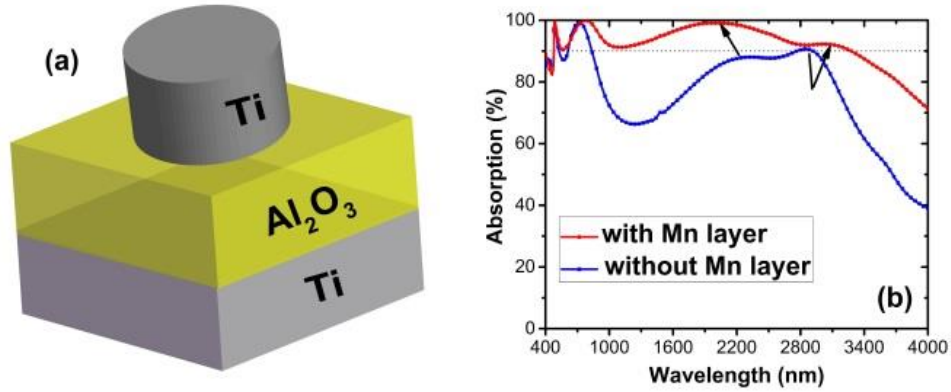


Figure 4.6: (a) 3D schematic of the unit cell of the structure without the Mn layer, and (b) absorption of the optimal structure with and without the Mn layer shown in red and blue colors, respectively.

To better illustrate importance of the 5 nm Mn layer, we investigate the optical response of the suggested structure without this layer, i.e. when Mn is replaced by Al₂O₃ and the remaining structure is a top-layer-patterned MIM configuration. The schematic of the structure without the ultra-thin Mn layer is presented in Fig. 4.6(a) (also see Fig. 4.1(d)). The absorption spectra of the structures with and without the Mn layer are shown, respectively, by red and blue lines in Fig. 4.6(b). The absorption band extends from 478 to 526 nm and from 610 to 836 nm for the configuration without the Mn layer, i.e. it is composed of two adjacent bands. Although the absorption spectrum has four peaks at the wavelengths of 482, 722, 2,320, and 2,848 nm in this case, the third peak at larger wavelength does not have a significant strength. Therefore, the resulting absorption band is relatively narrow (it comprises the above-mentioned adjacent bands) with the total bandwidth of 274 nm, in the case of not using the Mn layer. After adding the Mn layer, the strength and the spectral position of the first two peaks do not change significantly, but the strength of the third and fourth peaks considerably increases, and they also experience some shift that is depicted in Fig. 4.6(b) with the black arrows. This conveys that the first and second absorption peaks do not depend on the effect exerted by the Mn layer, while the third and fourth ones are significantly affected by the Mn layer. This is in total agreement with our observations in Figs. 4.5(a) and 4.2(d). Then, by adding the Mn layer, the bandwidth gets enhanced from 274 to 2,800 nm, i.e. it experiences a tenfold increase. It is worth

noting that adding an ultra-thin layer of Mn to the structure does not add considerable complexity to the fabrication process.

The absorption peaks may be due to different types of resonances such as LSP, PSP, or guided-mode resonance (GMR). The LSP resonances can occur because of the existence of nanodisks [36], the PSP mode can propagate in the continuous interface of the bottom Ti and Al_2O_3 layer [127], and GMR can appear in the Al_2O_3 layer that can work as a dielectric waveguide starting from a certain value of d_i [128-130]. It is noteworthy that, as follows from the results of simulations performed for different materials of dielectric (i.e. other than Al_2O_3) and its thickness, GMRs may provide the main contribution to the resulting mechanism behind the first peak, in some ranges of variation of the geometrical and material parameters, while PSPs do so for other ranges. Moreover, it has been observed that the GMR-related and the PSP-related effects may coexist in one structure at the same frequency, and sometimes it is difficult to distinguish between the extents to which each of them contributes. For instance, based on our simulations, the dominant effect of a PSP for the first peak is obvious in case of a thinner Al_2O_3 layer (e.g. at $d_i=80$ nm). In such a case, the field pattern is similar to the one in [127]. At the same time, as will be exemplified below for our optimal parameters, a GMR can dominate at larger thicknesses of the dielectric layer. As the full classification of the possible physical effects and an estimation of the extent of their contribution to the resulting absorption mechanism are beyond the scope of this section, we focus here on the optimal (main) structure.

In order to obtain some vision of the physical phenomena underlying the observed absorption peaks, the magnitude of magnetic field distribution inside the structure at the xz-plane cross section of the unit cell (see Fig. 4.1(c)) is presented in Fig. 4.7. Figs. 4.7(a-d) show the H-field pattern at the first, second, third, and fourth absorption peak's wavelength, respectively. Because of the symmetric nature of the designed structure, it is polarization insensitive for the case of normal incidence. Without any loss of generality, we may assume the electrical field vector of the normally incident wave to be in the x-direction, see Fig. 4.1(a). Therefore, since the propagation is in the z-direction, its magnetic field vector is in the y-direction. Field distributions in Fig. 4.7 show the y-component of the magnetic field (H_y) at the four peak wave-

lengths of the main structure. It is noteworthy that for this cross-section, the only component of magnetic field is H_y , while H_x and H_z are zero.

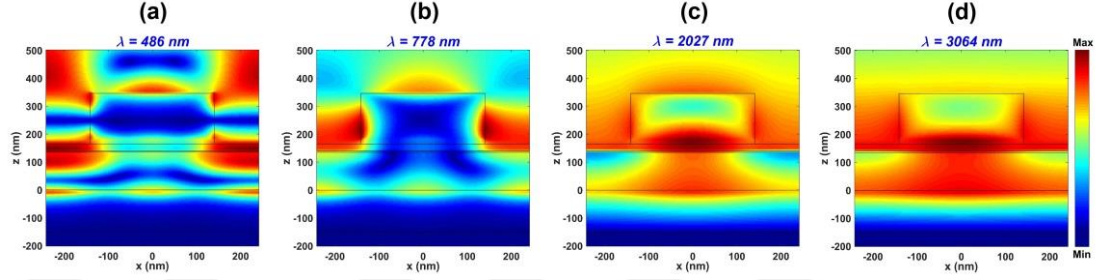


Figure 4.7: The spatial distribution pattern of the magnitude of the y-component of the magnetic field (H_y) in the xz cross-section of the structure at the (a) first, (b) second, (c) third, and (d) fourth absorption peak

It can be observed in Fig. 4.7(a) that, at the first peak of the main structure, the field is strong in the Al_2O_3 layer that indicates possible connection to a GMR. Moreover, some features are seen, which indicate possible contribution of PSPs at the bottom Al_2O_3 -Ti interface. At the second peak, the magnetic field distribution, which is shown in Fig. 4.7(b), indicates some localization around and between the nanodisks, so that the excitation of an LSP mode occurs here due to the coupling between the adjacent Ti disks. Field patterns of the third and fourth peaks shown in Figs. 4.7(c,d) are very similar to each other, and both are showing localization of the field under the nanodisks, inside the dielectric layer. For the fourth peak, the field pattern indicates the possible excitation of an LSP mode (or gap-plasmon mode) inside the dielectric gap between the underside of nanodisks and bottom Ti layer [17]. Moreover, a strong field enhancement can be observed around the ultra-thin Mn layer and inside it, which may appear due to optical properties of Mn (will be explained in-detail later), that allows the field to penetrate and transmit through the layer easily, and get absorbed inside it. This is why the absorption of the Mn layer is higher compared to the other two metallic components at large wavelengths (see Fig. 4.5(a)), and adding the Mn layer enhances the absorption of the entire structure rather at large wavelengths than at the small ones (see Fig. 4.6(b)). The situation is similar but not identical for the third peak. In this case, an LSP cannot exist because the real part of permittivity is positive for all of the used materials. In fact, we have an all-dielectric lossy structure in this case. Therefore, we rather have here the mimicking of an LSP

mode. It might be in qualitative coincidence with the recently suggested theory of surface-plasmon-like effects in all-dielectric structures [131], but it needs an additional clarifying.

To demonstrate possible effects of a GMR and an LSP mode, the absorption spectrum of a periodic array of Ti nanodisks on a 165 nm thick Al_2O_3 substrate is plotted in Fig. 4.8(a). The unit cell of the simulated structure is also shown in the inset of the figure and the dimensions are chosen as $p = 480$, $h = 180$, $r = 140$, $d_i = 165$ nm. Absorption peaks are observed at 535 and 775 nm, being a reminiscence of the first and second peaks of absorption in the main structure. The spectral location of the peak at 775 nm is so close to the second absorption peak of the main structure at 778 nm. It is because they both appear due to the excitation of a coupled LSP mode between the nanodisks, and in both cases, nanodisks are the same. In turn, the first resonance at 535 nm is associated with a GMR. The reason that its spectral position does not perfectly coincide with the first peak of the main structure is that in the case of GMR, a layer located below the Al_2O_3 layer affects the GMR wavelength. It can be shown that adding a back-side slab below the Al_2O_3 layer and changing the real part of its permittivity from unity to that of Ti leads to the gradual shift of the first peak closer and closer to the first peak in the main structure (as well as to the one in the main structure but without the Mn layer). This gives us more evidence that the first peak of absorption in the main structure is connected to a GMR.

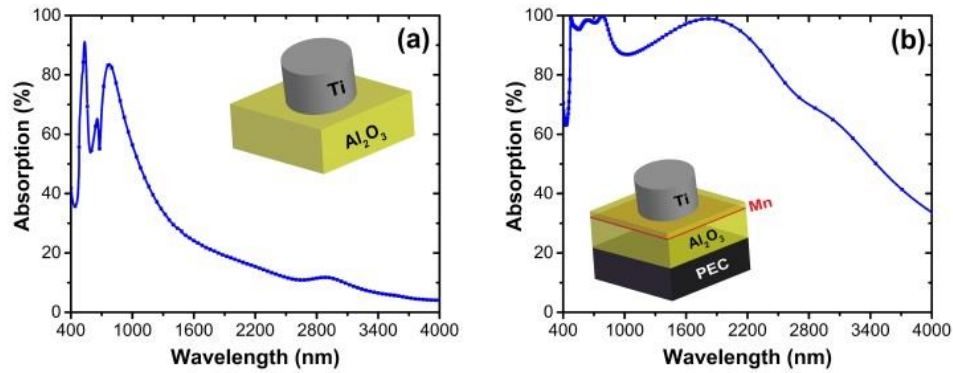


Figure 4.8: (a) Absorption spectrum of the periodic array of Ti nanodisks on the Al_2O_3 substrate, with $p = 480$, $h = 180$, $r = 140$, $d_i = 165$ nm. (b) Absorption spectrum in case when bottom Ti layer is replaced by PEC, with the same dimensions as those of optimized structure. The insets show the schematics of the simulated structures.

Next, Fig. 4.8(b) shows the absorption spectrum of the structure when the bottom Ti layer is replaced with a perfect electric conductor (PEC). The unit cell of the simulated structure is also shown in the inset of this figure, and the dimensions are the same as those of the optimized structure. This simulation is done to confirm the fact that the first absorption peak of the main structure is due to a GMR, and not a PSP mode. There are two absorption peaks at 477 and 783 nm, which almost perfectly match with the first and second absorption peaks of the main structure at 478 and 778 nm, respectively. This indicates that, in contrast with some of the earlier suggested absorbers [36, 127], these peaks are mainly not due to the excitation of a PSP mode at the bottom Al_2O_3 -Ti interface. Indeed, when there is a PEC at the bottom layer, any PSP mode cannot exist.

As we mentioned above, the field pattern of the first peak signifies the existence of a GMR. The phase-matching condition of the GMR in the 2D periodic structures is as follows [128, 129]:

$$k_{\text{GMR}} = k_0 \sin(\theta_0) + m \left(\frac{2\pi}{p} \right) \quad (4.1)$$

where P is the period of the structure, θ_0 is the incidence angle, K_{GMR} is the wave-number of the GMR, K_0 is the wavenumber of the incident wave in the free space, and m is an integer number. It is noteworthy that the condition given by Eq. (4.1) is also valid for PSPs in 2D periodic structures [36]. However, we confirmed above that the first peak of absorption for the main structure cannot be due to PSP. Eq. (4.1) conveys that by increasing (decreasing) the period of the structure, the spectral position of the GMR should experience a red-shift (blue-shift). To investigate this effect, the absorption spectra of the optimal structure with different values of p have been shown in Fig. 4.9(a). The range of wavelength is taken between 400 and 1,000 nm, for better evidence of the shifting behavior for the first two peaks. As expected, the first peak experiences a red-shift while increasing p . This confirms our earlier argument that the first peak is connected with a GMR. The second peak experiences almost no shift that indicates no connection to a GMR or PSP.

Let us remind here that the second peak is due to the excitation of a coupled LSP mode (Fig. 4.7(b)) and, fourth and third peaks are expected to appear due to the exci-

tation of a gap-plasmon LSP mode and the mimicking of such a mode in all-dielectric case, respectively (Fig. 4.7(c,d)). Figures 4.9(b) and 4.9(c) demonstrate the absorption spectra of the optimal structure for different values of r and h , respectively, with the wavelength ranging from 400 to 1,000 nm. It can be observed that the increase of r or h causes an obvious red-shift in the position of the second peak, while the position of the first peak is almost insensitive to any variation of r and h . The behavior of the first peak is already expected from its origin being associated to a GMR, and not an LSP mode, while the behavior of the second peak can be clearly explained from the field pattern in Fig. 4.7(b). It is obvious that, because of the field localization and strong confinement of the field in the region between the adjacent disks in the case of the coupled LSP mode associated with the second peak, both r and h should affect its spectral position. Moreover, the behavior of the third and fourth peaks by changing r and h is evident in Figs. 4.3(c) and 4.4(c), respectively. It is evident in Fig. 4.3(c) that increasing r significantly red-shifts both the third and fourth peaks, while the increase of h exerts a very minor effect on the third peak and almost no effect on the fourth peak (Fig. 4.4(c)). These features can also be explained by using Figs. 4.7(c) and 4.7(d). One can see that the magnetic field for the conventional and mimicked gap-plasmon LSP modes for the both peaks are below the nanodisks, i.e. inside the dielectric (Al_2O_3) region and, therefore, they are associated with the radius of disks, and not with their height.

The input impedance of the entire structure, which is normalized to the free-space impedance can be calculated by using the following formula:

$$Z_{in} = \left(\frac{(1 + S_{11}^2) - S_{21}^2}{(1 - S_{11}^2) - S_{21}^2} \right)^{0.5} \quad (4.2)$$

where S_{11} and S_{21} are the elements of the S parameter's matrix, corresponding to the reflection and transmission, respectively. The results are presented in Fig. 4.9(d). In order to have perfect matching with free space, the real part of the normalized input impedance of the structure must be 1, and its imaginary part must be zero. At the regions where these conditions are met to some extent, i.e. even an imperfect impedance matching may lead to minimizing the reflection and, thereby, maximizing the absorption. For instance, $Z_{in} = 1.016 - j 0.029$ at the spectral position of the second

absorption peak for the optimized structure, located at 778 nm, which is the cause of the strong absorption. The cause is the same for the third and fourth peaks. Now, let us discuss some features observed in Fig. 4.2 once more, by taking into account the results presented in Fig. 4.10. It is observable that changing the metal of the ultra-thin layer has a much more significant effect on the absorption of the structure compared to changing the metal of the top nanodisk and bottom metal layer. The significant difference between the performances of different metals as the ultra-thin middle metal layer is attributed to the fact that, as it was observed in the field patterns in Fig. 4.7, the field is enhanced around this thin layer for IR wavelengths, and the field penetration and absorption inside this layer gets significantly affected by the optical properties of the used metal. It is noticeable from Fig. 4.2(d) that Mn provides the best result when being used as the thin layer. The second best performance has the

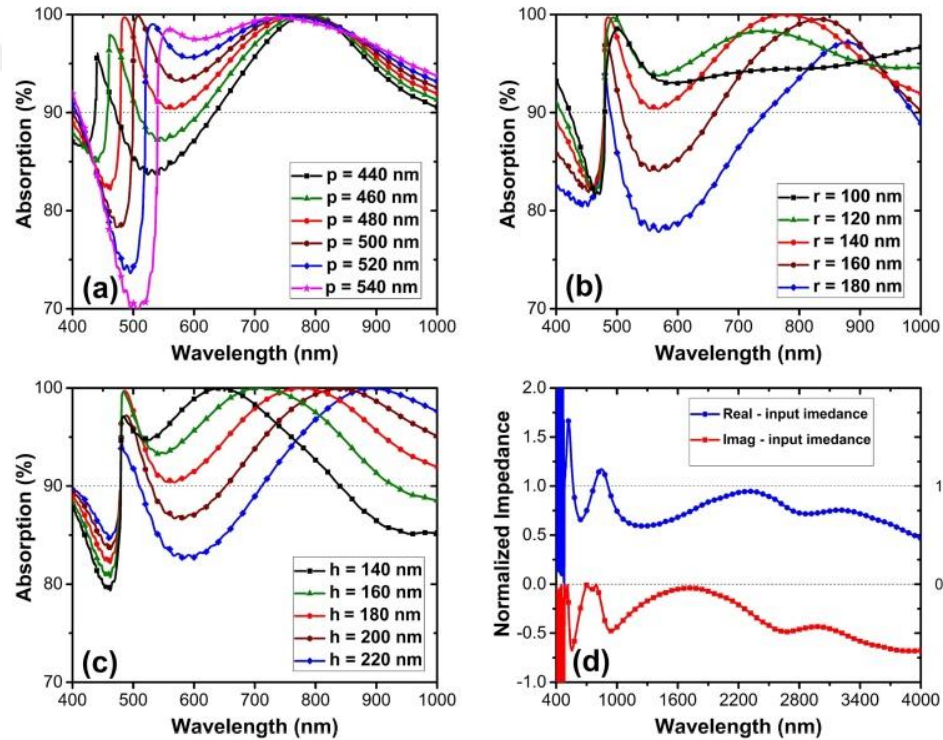


Figure 4.9: Absorption spectra of the optimal structure for different values of (a) p , (b) r , and (c) h ; (d) Real (blue line) and imaginary (red line) parts of the normalized input impedance of the optimal structure, Z_{in} , calculated using S-parameters, see Eq. (4.2).

ultra-thin layer made of Ti. These two materials of the ultra-thin layer provide a significantly superior performance as compared to the other metals in the MIR region. As seen in Fig. 4.10(a), Mn and Ti have much smaller modulus of the real part of permittivity compared to the other metals, in the MIR region. Having a small modulus of the real part of permittivity leads to a high field penetration inside Mn and Ti, and their intrinsic metallic loss may lead to stronger absorption. Mn has a smaller modulus of the real part of permittivity and a larger imaginary part of permittivity (higher loss), compared to Ti. This is why using Mn as the ultra-thin layer results in a wider absorption band compared to Ti. Moreover, it can be observed in Fig. 4.2(d) that using Au and Ag in the ultra-thin layer leads to a significant narrowing of the absorption band. This issue arises from the similar reason as the one explained above. It can be observed in Fig. 4.10 that Au and Ag have a much larger modulus of the real part of permittivity and a smaller imaginary part of permittivity compared to the rest of metals. This is a reason why the field penetration, field enhancement, and absorption for Au and Ag cannot be as efficient as for the other metals.

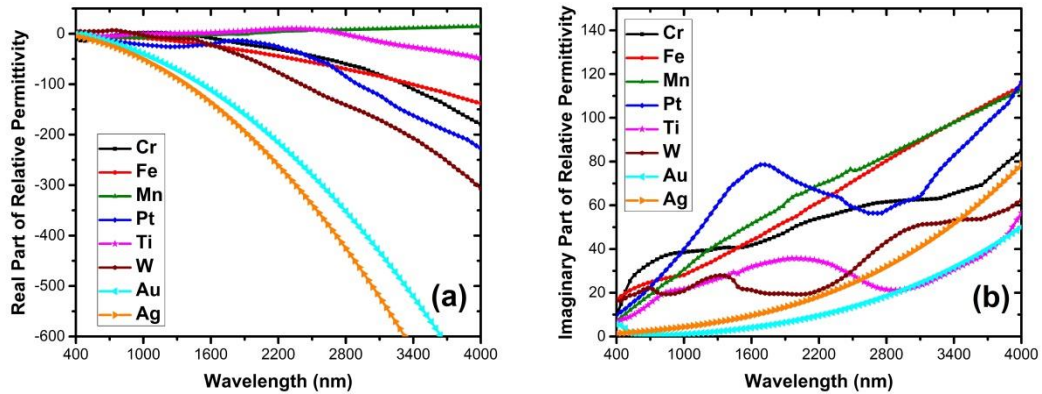


Figure 4.10: (a) Real and (b) imaginary parts of the relative permittivity for Cr, Fe, Mn, Pt, Ti, W, Au, and Ag.

4.3.3 Fabrication Tolerance

Next, let us investigate the fabrication tolerance of the structure, which is an important factor for the evaluation of the prospects of mass production. As mentioned earlier, large-scale fabrication and production of the proposed structure is realizable

by nanoimprint lithography, since it has only one step of lithography in its fabrication procedure. This technique can resolve sub-100 nm features with a high precision [39].

Figs. 4.11(a,b,c,d,e,f) show the color plots of the absorption versus wavelength for the continuously varying values of p , h , r , t_{Mn} , d_i , and d_{Mn} , respectively, while keeping all the other parameters constant and equal to the ones of the optimized structure. These figures demonstrate the tolerance of the structure's response to the deviations from the design dimensions, which can be a result of fabrication imperfections. The optimal values of the geometrical sizes are shown by the horizontal dashed lines. The response of the structure seems to be most sensitive to the deviations of the thickness of the Mn layer (t_{Mn}), as shown in Fig. 4.11(d). In particular, one can see that the deviations of t_{Mn} from the selected value can lead to a band narrowing (at larger t_{Mn}) and a band splitting (at smaller t_{Mn}). In addition, as can be seen in Fig. 4.11(c), deviations from the optimal value of r may lead to an insignificant red-shift or blue-shift in the position of the second, third, and fourth peaks, but the general behavior of the absorp-

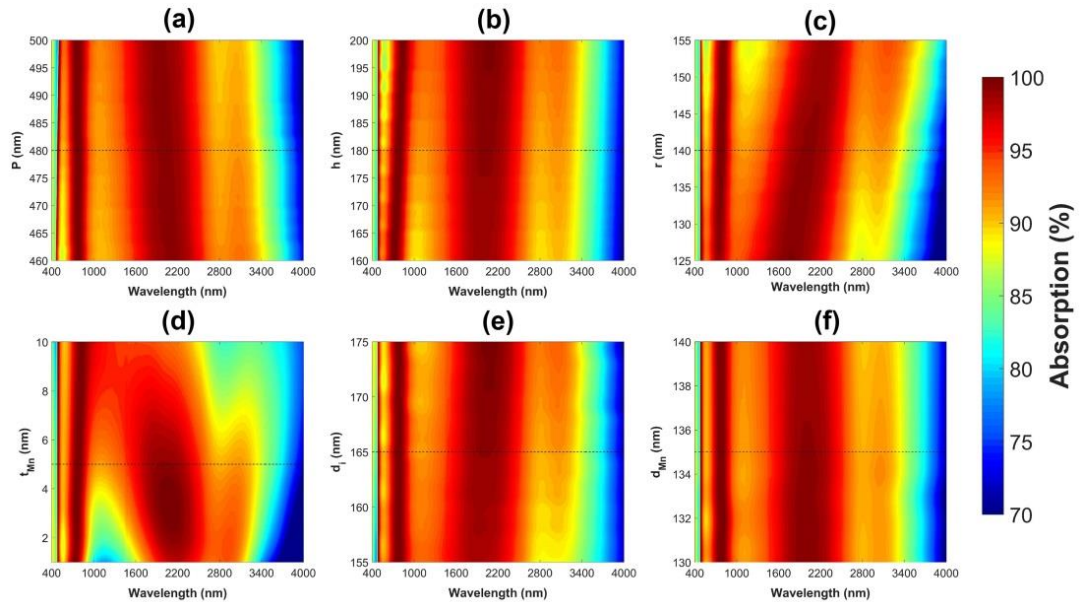


Figure 4.11: Color plots of the absorption versus (a) p , (b) h , (c) r , (d) t_{Mn} , (e) d_i , and (f) d_{Mn} and the wavelength, for the purpose of demonstration of fabrication tolerance of the optimal structure. The dashed horizontal lines indicate the optimal value of the parameter under study.

tion band does not get affected significantly. The response has a very good robustness to the introduced variations in p , h , d_i , and d_{Mn} , although some relatively minor changes in the bandwidth are possible. All the plots in Fig. 4.11 confirm that the suggested design has a very good fabrication tolerance to possible deviations from the optimal values. This confirms that, in addition to comprising cost-effective materials, this structure is fabrication tolerant and, therefore, it is a very good candidate for future mass production.

4.3.4 Polarization and Incidence Angle Sensitivity

As the final step, we investigate the absorption behavior of the structure for different angles of incidence for both polarizations. Figures 4.12(a,b) show absorption versus wavelength for incidence angles of 20, 40, and 60 degrees for TM and TE polarizations, respectively. In general, the absorption band is narrowing when the angle is increased. However, at certain wavelengths, for the case of the incidence angle of 40° and TM polarization in Fig. 4.12(a), absorption is almost perfect, e.g. $A > 97\%$, within the range of $818 < \lambda < 2,147$ nm. The results show that the absorption remains high in a wide wavelength range for TM polarization up to incidence angles as large as 60 degrees; TE polarization is more sensitive to the incidence angle variations. In spite of this, rather wide bands of $A > 90\%$ can exist, at least up to 40 degrees.

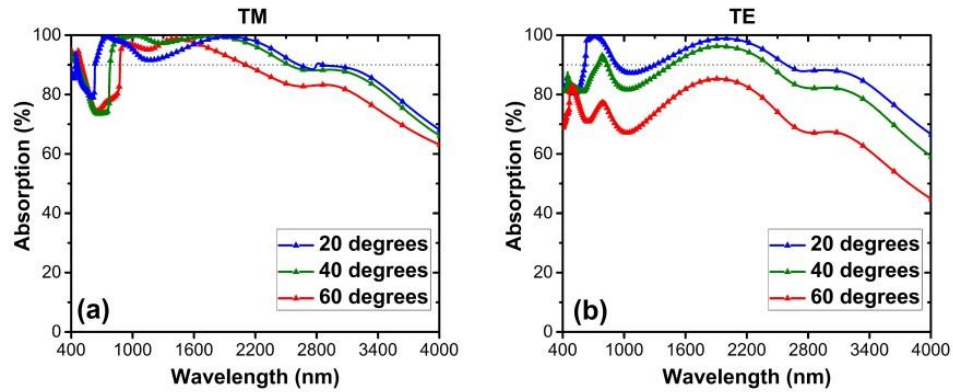


Figure 4.12: Absorption spectrum of the optimal structure for oblique incidence angles of 20, 40, and 60 degrees; (a) TM and (b) TE polarizations.

4.4 Conclusion

An ultra-broadband, nearly perfect absorber is designed, analyzed, and characterized in detail. The studied structure is actually a top-layer-patterned MIM structure with Ti-Al₂O₃-Ti configuration, with an ultra-thin 5 nm layer of Mn embedded into the dielectric (Al₂O₃) layer. It has been shown that adding just a very thin layer of Mn widens the bandwidth from 274 nm to 2,800 nm, i.e. it leads to a tenfold increase in the bandwidth. This allows covering the visible, NIR, and partially MIR range, while it does not need additional lithography steps and does not add much to the overall fabrication complexity of the structure. The structure has the relative absorption bandwidth of 149%, which is probably the best result obtained so far for such structures. It has been shown that the proposed structure keeps high absorption for both TM and TE polarizations up to large angles. Since the structure only needs one step of lithography in its fabrication procedure, with the dimensions of lithographic patterns being over 180 nm, and with the high fabrication tolerance, its mass production can be easily realized by using nanoimprint lithography technology. Taking this fact into consideration along with the unprecedentedly ultra-broadband, nearly perfect absorption, one can conclude that the proposed absorber is very promising for photovoltaic and thermal emission applications. The use of Mn for one of the absorber components, the used design procedure, and obtained numerical results can serve, along with the physical performed analysis, as a good starting point for design of a wide class of ultra-wideband absorbers that meet the requirements of a broad spectrum of applications, ranging from photovoltaics and solar cells to shielding and optical communication filters.



Chapter 5

Manganese-Based Lithography-free UV to FIR Absorption

5.1 Introduction

In this section, we present a facile and straightforward approach for the fabrication of ultra-broadband lithography-free absorbers by a single film of Mn coated over a high-roughness substrate including randomly sized needle-like and sharp nano-pyramids. The measured absorption using an integrating sphere confirms an average of 99% absorption in the ultra-broad range of 250-2500 nm, i.e. from UV to MIR. Simulations predict that the absorption above the 90% regime should at least go above 10 microns, which means UV to FIR. It is explained that such a broadband absorption is due to the cooperative contribution of two key factors: 1. electromagnetic field tapering owing to the tapered nature of the structure (graded-index or moth-eye effect), and the very appropriate optical properties of Mn for broadband absorbers [114, 120]. These physical characteristics contribute to the fact that the coated Mn film surprisingly, not only blocks the transmission but also suppresses the reflection. Such a result, to the best of our knowledge, is the best absorber realized with lithography-free structures. This concept can also be generalized to high-roughness substrates with different materials because, as mentioned, the field does not penetrate below the Mn layer and, therefore, the substrate only is required to have tapered features on it, independent of the type of its material. Such a desirable result with a relatively simple and inexpensive fabrication process can be an excellent candidate for mass

production purposes with a variety of applications ranging from photovoltaics to thermal emission and shielding.

5.2 Results and Discussion

Planar metal films with a thickness sufficiently larger than their skin depth are used as the bottommost part of the broadband absorbers to guarantee almost zero transmission. However, the same metal film, if used solely, leads to a high reflection due to its large impedance mismatch with free-space. Therefore, absorbers typically include other layers above the metallic substrate that suppress the reflection by providing impedance matching with the free space. In this section, we use only one metallic thin film of the thickness of 600 nm coated on a substrate with tapered features to obtain transmission and reflection suppression at the same time in an ultra-broad range, and thus reach an ultra-wideband absorption. This design does not include any other layers or materials on top of this metallic film.

Figure 5.1(a) shows the plots of transmission (T), reflection (R), and absorption (A) for a 600 nm planar thin film of Mn. The absorption can be calculated as $A=1-R-T$. It can be observed that, as expected, a metallic layer with such a thickness leads to almost zero transmission, but the reflection is large, and thus the total absorption is low. The idea in this work is to coat the same type of metallic single thin film but to make it in a tapered configuration that enables the moth-eye or graded-index effect. Figure 5.1(b) shows the simulated R, T, and A plots for the case of making the thin film of Mn in a periodic pyramidal configuration with the thickness of 600 nm, where the period is 1 micron and the height of pyramids is 4 microns. In other words, we have suspended periodic pyramids made of a 600 nm thick Mn layer. The schematic of such a case is shown in the inset of Fig. 5.2(b). We note that, by making this thin film tapered, in addition to the suppression of T, R gets suppressed in an ultra-broad range of wavelengths. This leads to having perfect absorption in the broad band of UV to FIR. The physics behind this broadband absorption will be elevated in detail in the next sections. It is also noteworthy that the permittivity of Mn (and Si) used throughout this section and in the simulations is measured by fitting measured spec-

troscopy data obtained by J. A. Woollam Co. Inc. V-VASE and IR-VASE ellipsometers.

In order to experimentally implement the above-mentioned idea, we should have a substrate with tapered features on it to coat the Mn layer. For instance, one can pattern a substrate with the shape of periodic nano-pyramids using EBL or FIB, but these methods are not preferred for scalable production, are expensive, very time-consuming, and they do not provide us with large-area structures. To avoid such a limitation, we use a lithography-free method that leads to randomly-sized nano-pyramids on the surface of a commercial 4-inch p-type Silicon (Si) wafer. Randomly-sized nano-pyramids on the surface of a commercial 4-inch p-type Si wafer are generated by using an inductively coupled plasma (ICP) tool and a Bosch process like etching recipe [132]. The use of etching to obtain nano-pyramids on the surface of an Si wafer is carried out in the previous works with other recipes and leads to the so-called Black Silicon which has applications as broadband absorber or solar cell. However, as will be experimentally presented in the coming sections of this study, absorption in Black Si is limited up to around 1000 nm because of the limited absorption properties of the doped Silicon [133]. However, as will be shown in the next sections, any substrate made of any other material that has tapered features on it leads to the same result. In other words, the ultra-broadband absorption of the structure is not due to the properties of Si and is because of the tapered geometry and optical properties of the Mn layer.

Figure 5.1(c) shows the schematic of the randomly sized and randomly distributed nano-pyramids on the surface of an Si wafer before and after coating with Mn. In order to achieve such features on the surface of the Si substrate, we have adopted an inductively coupled plasma (ICP) tool process recipe with multiple cyclic steps that are composed of deposition and etch steps, using the STS Multiplex ICP Etch system. Each cyclic step consists of two half steps. The first half step is reactive ion etching (RIE) of Si and the second half step is the deposition of C_4F_8 on the previously RIE etched Si surface. In the first half step (RIE), a mixture of SF_6 and O_2 gases are released into the process chamber for a duration of 10 seconds. In the second half step (deposition), C_4F_8 is deposited on the Si surface for 7 seconds. Gas flow rates for SF_6 , O_2 , and C_4F_8 gases are set as 80 sccm, 5 sccm, and 70 sccm, respectively. The chamb-

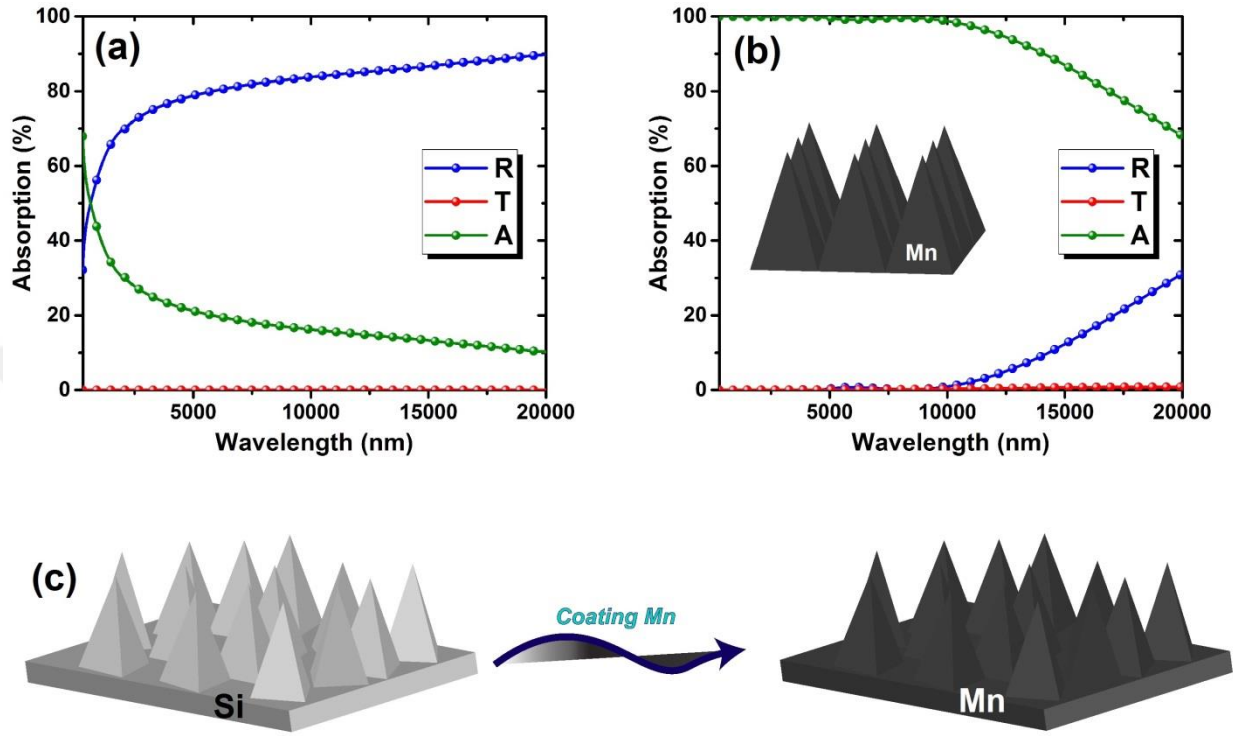


Figure 5.1: a) Transmission, reflection, and absorption spectrum of a 600 nm thick *planar* Mn layer, b) Transmission, reflection, and absorption spectrum of a 600 nm thick *suspended pyramidal array (shell)* of Mn, and c) schematic of the random Si nano-pyramids before and after coating the surface with e-beam evaporated Mn *shell like* thin-film.

er pressures during the half steps of etching and deposition are set as 35 mTorr and 20 mTorr, respectively. During the etching and deposition half steps, the coil power is set as 500 Watts and 400 Watts, respectively, and the platen power is set as 13 Watts and 0 Watts, respectively. The chamber temperature is set to 20°C for both of the half steps and, therefore, the entire process. Table 1 in supporting information shows the process recipe parameters for both of the deposition and etching half steps.

Figure 5.2(a-d) demonstrate the Scanning Electron Microscopy (SEM) images of the surface of a commercial p-type silicon wafer substrate when 100, 200, 300, and 400 cycles of the above-mentioned ICP tool recipe is applied on it, respectively. It can be observed that with 100 cycles of etching, the tip of the random nano-pyramids starts to appear, but there are still significant flat spaces on the substrate surface. For

the cases of 200, 300, and 400 process cycles, the nano-pyramids have obviously appeared and as expected, the height of pyramids increases as the number of cycles increase. In other words, the etching depth increases with the number of cycles. It can be observed in Figs. 5.2(b-d) that the shaped nano-pyramids are sharp and needle-like and they randomly vary in their size and distribution. The flat areas without tapered features on the surface of the 100-cycle etched wafer make it inappropriate for our purpose. Therefore, we coat Mn on the surface of the other three samples with 200, 300, and 400 etching cycles. Using the above-mentioned recipe, we obtained such features on a full 4-inch Si wafer and, therefore, in principle, this process is wafer-scale and can be applied to any size of wafers. However, the process parameters may need to be modified if larger scale wafers (i.e. 6-inch diameter, or 8-inch diameter Si wafers) or smaller scale wafers (i.e. 2-inch diameter Si wafers) are used instead of 4-inch diameter Si wafers.

The measured absorption spectra of the samples, the SEM images of which are shown in Figs. 5.2(a-d) are demonstrated in Fig. 5.2 (e), along with the absorption spectrum of the commercial Si wafer. It can be observed that, as expected, the 100-cycle sample does not give high absorption and has an absorption spectrum close to that of Si wafer before etching. On the other hand, all other three samples with 200, 300, and 400 etching cycles show very high absorption in the range up to around 1000 nm wavelength, and their absorption profiles are very similar to each other. The absorption of these samples drops significantly after 1000 nm, which is due to the reduction in the loss of the Si wafer. The images of the 100-cycle and 400-cycle etched Si full wafers are shown in Figs. 5.2(f) and 5.2(g), respectively. The black color of the 400-cycle sample is attributed to the high absorption of the nano-pyramids of doped Si in the visible region. However, as seen in Fig. 5.2(e), the absorption in this case is limited up to around 1000 nm wavelength, and for larger wavelengths, the structure loses its high absorption behavior. On the other hand, the 100-cycle sample has a non-black color close to that of commercial wafer without etching due to weak absorption.

Here, we emphasize that in order to obtain the absorption (A) spectrum, we should measure the transmission (T) and reflection (R). T can be measured by using normal devices such as ellipsometer or FTIR. However, for measuring R for this structure, we require an integrating sphere set up to be integrated with the device we are using.

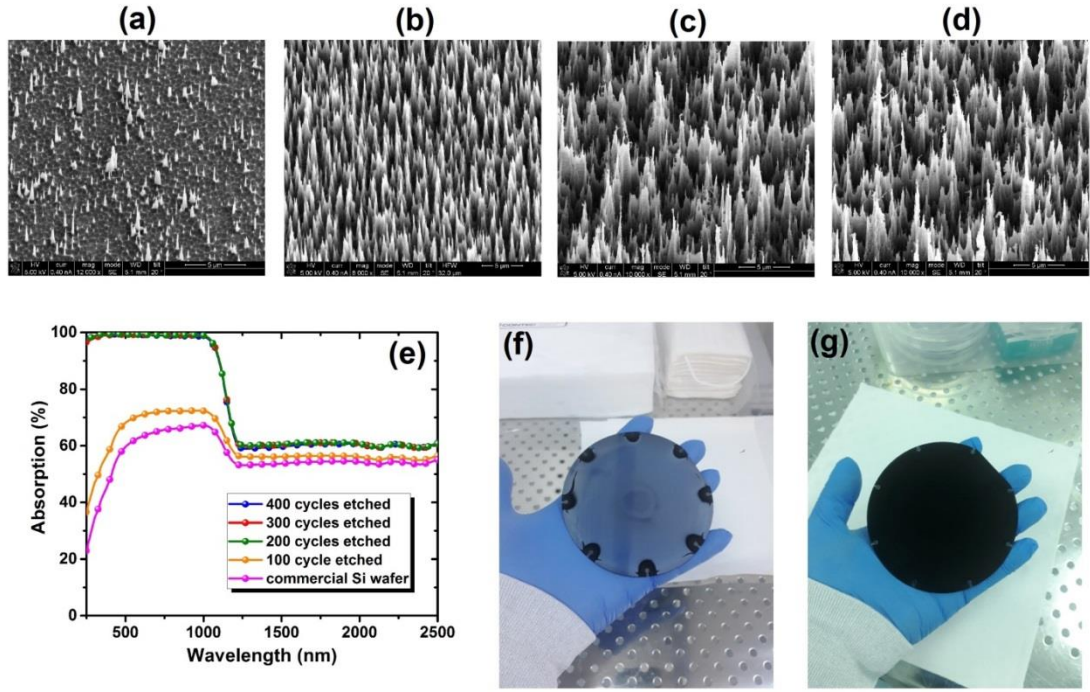


Figure 5.2: SEM image from the surface of the Si wafer etched for a) 100, b) 200, c) 300, and d) 400 process cycles. e) Absorption spectra of the samples mentioned in (a), along with commercial Si wafer. Photograph of the Si wafers etched for f) 100, and g) 400 cycles.

This is due to the fact that, because of the nonplanar geometry of the structure, a high portion of the reflected wave will be scattered, i.e. we have to measure both diffusive and specular reflections from the sample. Any device without an integrating sphere setup only measures the specular reflection. However, the integrating sphere has a reflective mirror inside it and gathers all portions of the reflected beam which is scattered into any direction and sends all to the detector. The integrating sphere used in our device, is attached to the Carry 5000 UV-VIS-IR spectrophotometer and measures in the range of 250-2500 nm. The sphere is covered with PTFE and it is 110 mm in diameter. The transmission measurements are carried out using V-VASE and IR-VASE Ellipsometers.

In order to extensively broaden the perfect absorption band, we coat a single layer of Mn with three different thicknesses of 400, 600, and 800 nm, using VAKSIS electron-beam evaporator system. The coating rate of Mn is set to 10 Angstroms/s. The relatively high rate of coating is due to the fact that in this work it is not required to

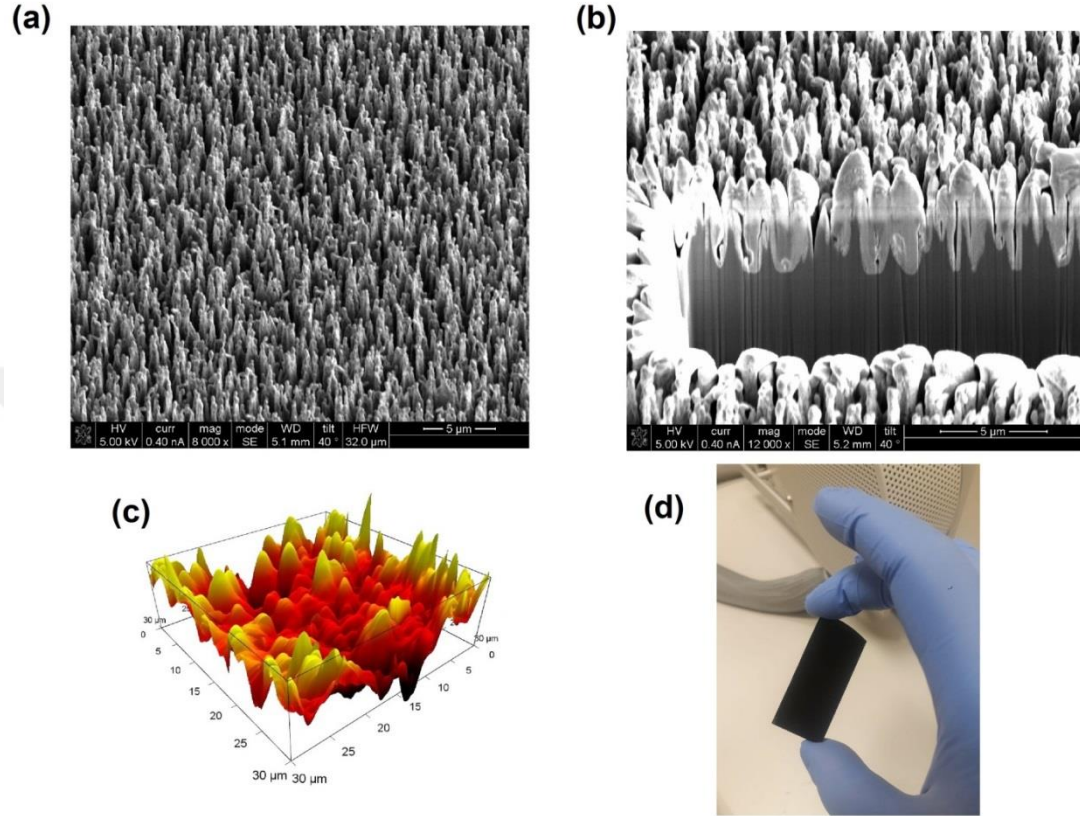


Figure 5.3: a) SEM image of the surface of the 300-cycle etched sample coated with 600 nm Mn, b) SEM image of the cross-section of the sample mentioned in (a) that is milled by FIB, c) AFM image of the surface topography of the sample mentioned in (a), and d) photograph of the sample mentioned in (a).

have flat and conformal films and, therefore, we can use high coating rates which is very desirable for high-throughput production purposes. On the other hand, in broadband absorption applications, having extra roughness and non-conformal layers generally leads to a broader absorption band since these features may add extra resonances to the overall performance of the structure.

The SEM image of the 300-cycle sample coated with 600 nm Mn is shown in Fig. 5.3(a). Moreover, Fig. 5.3(b) demonstrates the cross-section SEM image of the same sample which is milled by Focused Ion Beam (FIB). From Fig. 5.3(b) it can be observed that the randomly sized Si nano-pyramids are coated with Mn and the thickness of Mn layer is not uniform. More images from the cross-section of the same sample can be found in supporting information. Fig. 5.3(c) also shows the Atomic

Force Microscopy (AFM) image of the surface topography of the sample. Finally, Fig. 5.3(d) shows the image of the mentioned sample.

Figures 5.4(a), (b), and (c) show the absorption spectrum for the 200, 300, and 400 cycles etched samples, respectively. Each figure contains the measured absorption for the three thicknesses of coated Mn, 400 nm, 600 nm, and 800 nm. The variation of etching cycles and coated Mn thickness is to check the possible changes of the absorption spectrum's behavior with regards to the fabrication parameters. It can be observed that, throughout the whole measured wavelength range of 250-2500 nm, the absorption is almost perfect for all of the fabricated devices, and their performances are almost the same. In other words, the measured absorption confirms perfect absorption ranging from UV to MIR with negligible dependence to variations in the fabrication process, and the structure shows an excellent fabrication tolerance. The average absorption in the measured range is approximately 99%. Such wideband absorption with a lithography-free structure has not been experimentally reported before, to the best of our knowledge. It should also be noted that the integrating sphere setup is limited up to the wavelength of 2500 nm. As it will be shown in the following, simulations confirm that the absorption band of the structure goes up to around 10 microns, i.e. from UV to FIR. It is also evident from the trend of measured absorption behavior of the structure in Fig. 5.4 as the wavelength increases, that the absorption does not show a negative slope toward the larger wavelengths, and if the same trend continues for the larger wavelengths that have not been measured, the absorption will remain above 90% for up to a couple of microns larger than 2500 nm wavelength. It is noteworthy here that, as it will be confirmed by the simulations, the high-roughness Si wafer does not contribute to blocking the transmission, and is almost completely blocked by the Mn layer that in all cases is a couple of hundred nanometers. The color of 600 nm thick Mn coated 300-cycle etched sample, the image of which is shown in Fig. 5.3(d), and all of the other samples are black due to the perfect absorption of the visible light. The objective of fabricating 9 samples with 3 different etching cycles and 3 different Mn thicknesses is to show that this method has a very high fabrication tolerance and the combination of tapered substrate with the optical properties of Mn is the key to this broadband absorption and it is not restricted to any specific geometrical optimization. As demonstrated with the experiments, having high fabrication tolerance is a very important feature for the mass production of such absorbers.

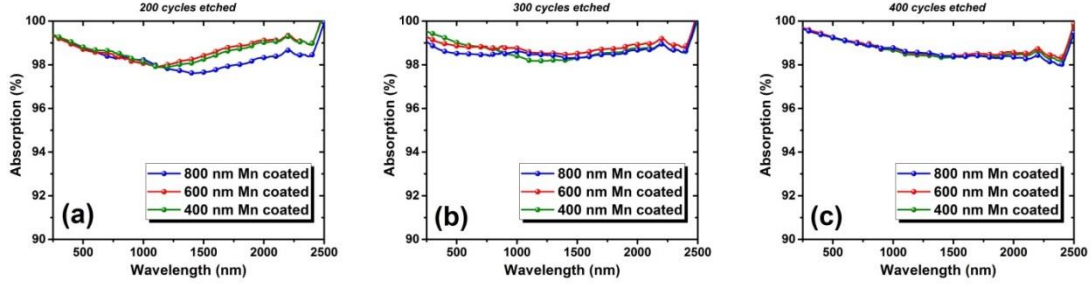


Figure 5.4: Absorption spectra of the fabricated samples with the three different Mn coating thicknesses of 400 nm, 600 nm, and 800 nm, for the cases of a) 200, b) 300, and c) 400 cycles of etching.

Based on the topographic image of the sample (Fig. 5.3(c)), we can approximately model the 300-cycle 600 nm Mn coated structure as a periodic array of Mn coated nano-pyramids. The 3D schematic of the approximated model of the structure, along with the xz-plane cross-section of its unit cell is shown in Fig. 5.5(a). In this approximation, the whole structure is assumed to be made of similar periodic pyramids. As can be seen in Fig. 5.5(a), the height of the inner pyramid made of Si, the period, and the thickness of the coated metal layer are denoted as h , p , and t , respectively. Based on the SEM images taken from the cross-section of the sample, we choose $h=4$ microns, $p=1$ micron, and $t=600$ nm for the simulation. In this unit cell, the Si pyramid is not extended downwards below the region where Mn is coated. This is done to make sure that the transmission blockage behavior of the structure is not due to the high thickness of the Si wafer below the patterns and is only achieved by the Mn layer.

The absorption of the approximated structure is shown in Fig. 5.5(b). The simulations are carried out with two independent methods using the Lumerical for FDTD (Finite Difference Time Domain) [113] and COMSOL Multiphysics for FEM (Finite Element Modelling) [134]. It can be observed that such a structure shows an absorption of above 90% up to the wavelength of 13500 nm, i.e. spanning from UV to FIR. To discuss the results more clearly, the simulated reflection and transmission of this sample are also shown in the same plot. It can be observed that in the mentioned ultra-broad range, both the reflection and transmission are suppressed. The transmission is suppressed due to the fact that the thickness of the Mn layer is 600 nm which is much larger than its skin depth, therefore, this simulation proves that in the experime-

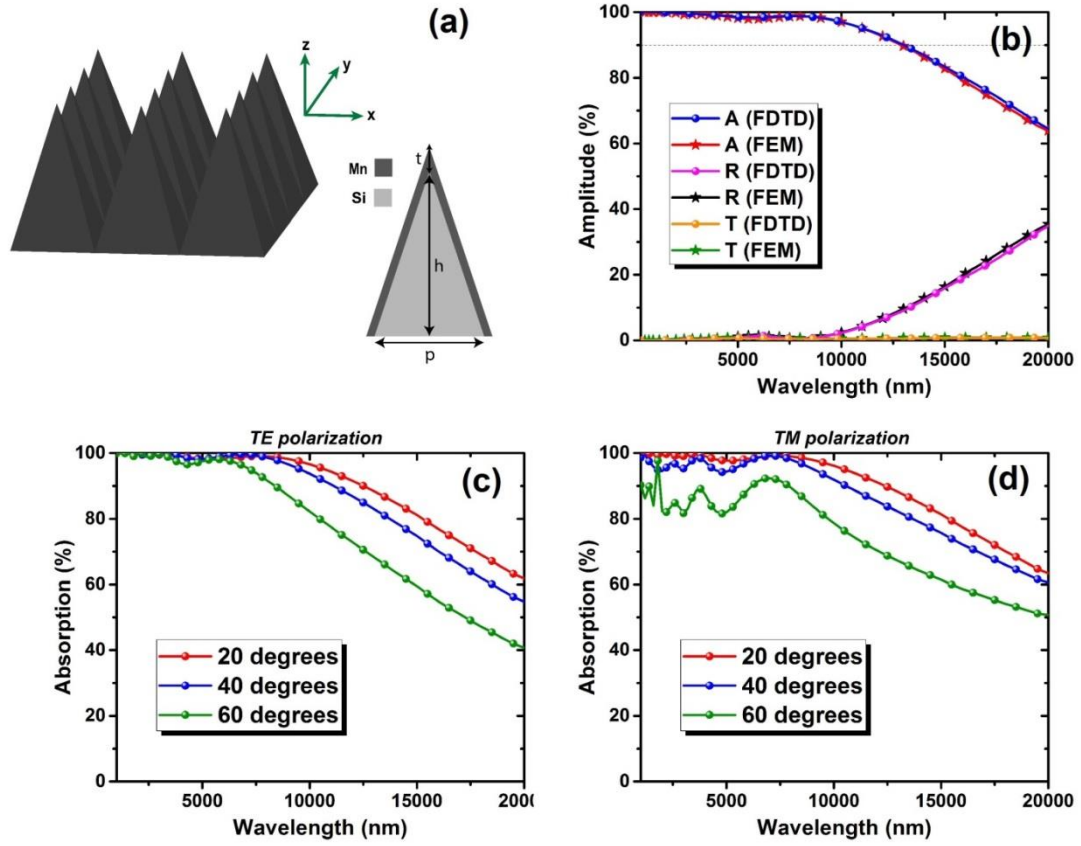


Figure 5.5: a) 3D schematic of the approximated model along with its xz cross-section of its unit cell, b) Calculated reflection, transmission, and absorption spectrum of the structure when $h=4$, $p=1$, and $t=0.6$ microns. Calculated absorption spectra for the incidence angles of 20 degrees, 40 degrees, and 60 degrees, for the case of c) TE polarization, and d) TM polarization.

ntal case, the transmission is not blocked as an effect of high thickness of the Si wafer below the nano-pyramids and is a direct result of the thickness of the Mn film only. The conclusion that thick Si substrate does not affect the results also enables us to fabricate the same structure on top of any high-roughness substrate other than Si, and shows that the absorption behavior of the structure is independent from the choice of substrate as long as the surface of the substrate has enough roughness. We further demonstrate this point by studying the field profile within the structure.

We note that the reflection is suppressed because of the effect of Field Tapering or the graded-index (moth-eye) effect. In the conventional structures, the metallic layers with thicknesses of a couple hundred nanometers are put at the bottommost part to

only block the transmission, and the minimization of reflection happens due to the contribution of the other parts of the structure above that bottom layer. However, unlike those conventional absorbers, in our case, a single coated metallic layer happens to play the role of blocking both the transmission and reflection at the same time. This phenomenon is due to the tapered geometric shape of the substrate that enables the effect of field tapering. This effect, along with the very appropriate optical properties of Mn and the light trapping nature of this structure, which will be discussed in detail later, contributes to such a broadband absorption.

Since it is not possible to use an integrating sphere setup for the case of oblique incidence, the same approximated structure is also simulated for the cases of TM and TE polarizations of oblique incidence. The absorption spectra for incidence angles of 20 degrees, 40 degrees, and 60 degrees and for TE and TM polarizations are presented in Figs. 5.5(c) and 5.5(d), respectively. It can be observed that the structure retains its high absorption for large incidence angles and for both polarizations, although it is more robust for the case of TE polarization.

Now, let us discuss the physical mechanism behind this broadband absorption in more detail. As the first step toward the physical analysis, the electric field magnitude pattern is shown at the xz-plane cross-section of the unit cell in Fig. 5.6. The simulations are carried out for the different wavelengths of 2, 4, 6, 8, 10, and 12 microns. The field patterns clearly demonstrate the phenomena of the field tapering due to the graded-index nature of the structure which is also referred to as the moth-eye effect. Field tapering happens when there is a gradual change in the effective index of the structure from the top to the bottom which leads to a slow and smooth impedance matching between the air and the structure. Here, obviously the structure-to-air ratio at any horizontal cross-section of the structure increases gradually which is the cause of graded-index effect in this pattern. This shows that the incident field slowly gets coupled to the structure in the region between the adjacent unit cells, but it cannot penetrate inside the coated nano-pyramids due to the high thickness of the Mn layer. After coupling slowly to the structure, the field gets absorbed by the lossy Mn layer.

Here, we demonstrate why Mn is an appropriate metal for this structure. Figure 5.7(a) shows the absorption spectrum for different coated metals in the configuration of

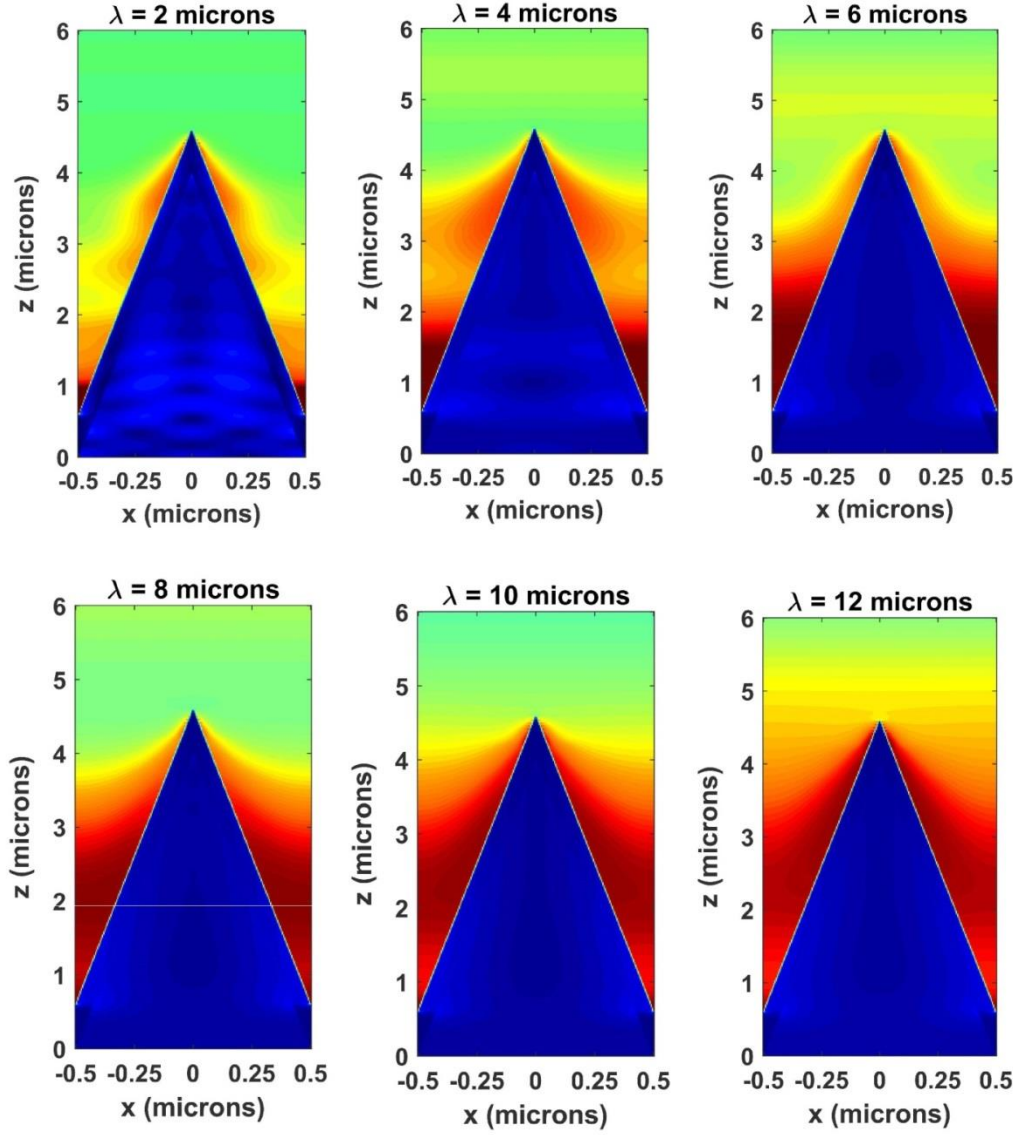


Figure 5.6: Electric field magnitude distribution at the xz cross-section of the unit cell for different wavelengths of 2, 4, 6, 8, 10, 12 microns.

Fig. 5.5(a), which are typically used for broadband absorber structures. These metals include W, Cr, Ti, Fe, and Mn. The dimensions are same as in the earlier simulations ($h=4$, $p=1$, and $t=0.6$ microns). It can be seen that for the case of other metals, the $A>90\%$ band at the best goes up to the wavelength of approximately 4.5 microns for the case of Ti, while when using Mn the bandwidth is enhanced very significantly and the upper wavelength limit of $A>90\%$ band goes up to around 13 microns. This significant superiority of Mn can be explored in its optical properties. For this purpose,

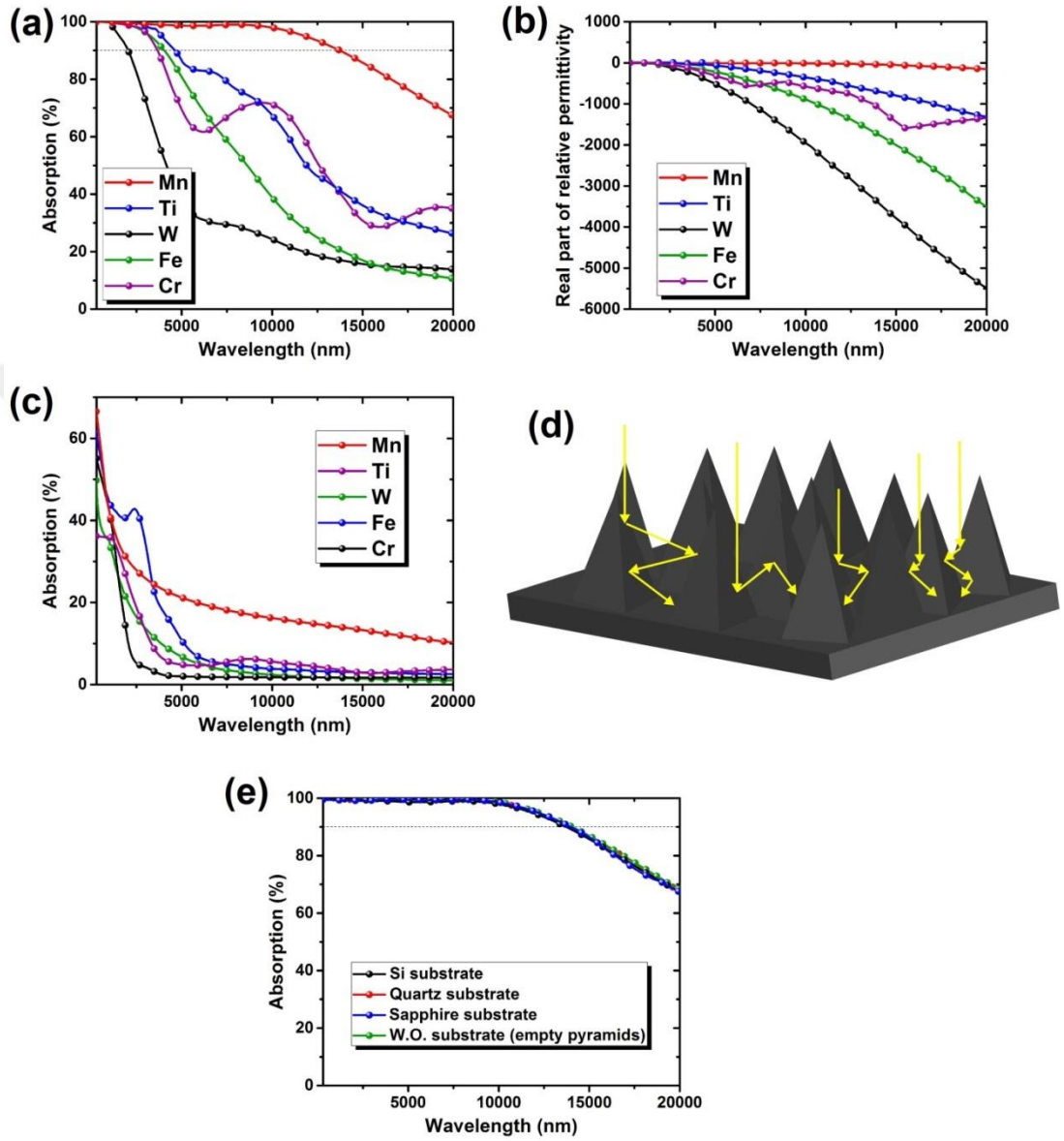


Figure 5.7: a) Absorption spectrum of Si nano-pyramid array coated with Mn, Ti, W, Fe, and Cr, b) real part of relative permittivity, and c) absorption spectrum of 600 nm thick planar film of the metals mentioned in (a), d) Schematic of the light trapping nature of the structure, and, e) absorption spectrum of the nano-pyramid array of different substrates when coated with Mn.

we have plotted the real part of relative permittivity of all the metals in Fig. 5.7(b). The optical data for Mn is obtained by ellipsometric measurements and for all other metals it is taken from Palik [123]. It can be observed that in such a broad wavelength range, Mn has a much smaller absolute value of real part of permittivity com-

pared to the other metals and it retains its small value throughout the whole spectrum. Having a small real part of permittivity leads to better field penetration inside the metal and through the intrinsic loss of metal, i.e. the imaginary part of permittivity, the penetrated field gets absorbed. Moreover, the absorption spectra of 600 nm planar films of the above-mentioned metals are shown in Fig. 5.7(c). It can be observed that even a planar film of Mn has a better absorptive capability compared to other metals.

The other physical mechanism (especially in shorter wavelength where ray optics approximation is applicable) contributing to the excellent performance of the proposed device is the light-trapping nature of the structure. It is common to trap the light inside Fabry-Pérot or gap-plasmon cavity modes, but in this structure, the non-planar configuration of the constituting elements makes it a nice scaffold for light trapping. As shown by the schematic presentation in Fig. 5.7(d), the light can get reflected back and forth from one surface to another between the neighboring pyramids, and in each reflection, it can be partially absorbed.

Based on the above-mentioned arguments, it can be summarized that the main physical mechanisms behind this broadband strong absorption are: 1. The phenomena of field tapering that exists due to the graded-index profile of the structure, and, 2. The significantly small real part of permittivity of Mn that allows for better field penetration inside the Mn film and consequently leads to higher absorption compared to conventional absorbing metals, and 3. The special geometry of the structure that makes an appropriate platform for light trapping.

As a final remark, the absorption spectrum of the structure for the case of using different substrate materials in the unit cell of Fig. 5.5(a) is shown in Fig. 5.7(e). This is done in order to show that this approach is valid to any high roughness substrate and is not a result of properties of Si. The other possible typical substrates are quartz (SiO_2), and Sapphire (Al_2O_3). The absorption spectrum of the structure for the case of not using any substrate material, with suspended empty pyramids is also shown in the figure. It is obvious that the result is almost robust to the choice of substrate material and the ultra-broadband absorption profile is almost the same for all the substrate materials, as well as, for the case of suspended pyramids. In other words, the extremely broadband absorption of this structure is obtained due to the geometric properties of

the substrate and is not connected to the substrate material. Therefore, the same result is obtainable by any other substrate material as long as the substrate has tapered pyramidal features on it.

5.3 Conclusion

In summary, a facile and feasible lithography-free approach for obtaining ultra-broadband perfect absorption through a single layer coating of Mn over a high-roughness substrate including densely packed and randomly distributed nano-pyramids, is experimentally presented. The experimental measurements using integrating sphere confirm approximately 99% average absorption in the range of 250 to 2500 nm, i.e. from UV to MIR. The measurements are limited to this wavelength due to the limitations of the integrating sphere. However, the simulations predict that the absorption band extends up to above 10 microns, in other words, from UV to FIR. The physical mechanism behind this broadband absorption is discussed and it is shown that this is due to the cooperative contribution of the superior optical properties of Mn along with the phenomena of electromagnetic field tapering and light-trapping between the densely packed and randomly distributed nano-pyramids. The tapered structures on the surface of the Si substrate are obtained by a Bosch process like recipe etching in an ICP tool. It is important to emphasize that, as demonstrated through the simulations, this approach can be used for any high-roughness substrate other than Silicon with densely packed, randomly distributed nano-pyramidal shaped features on the substrate. By fabricating several samples with different etching cycles and Mn layer thicknesses, and by achieving almost the same result from all of them, it is also experimentally shown that the structure has a high fabrication tolerance that makes it a very appropriate candidate for mass production. The findings and experiments of this section are very beneficial for large-area and mass production of broadband absorbers covering the full range of UV to FIR.

Chapter 6

Lithography-Free Electrically Tunable Color Filters

This chapter is on part reprinted with permission from: **Majid Aalizadeh**, A. E. Serebryannikov, A. Khavasi, G. A. E. Vandenbosch, and E. Ozbay, “Toward Electrically Tunable, Lithography-Free, Ultra-Thin Color Filters Covering the Whole Visible Spectrum,” *Scientific Reports*, vol. 8, p. 11316, 2018.

6.1 Introduction

In this work, we propose an electrically tunable, lithography-free, ultra-thin color filter. It is designed by utilizing the EO dielectric organic crystal of 4-dimethyl-amino-N-methyl-4-stilbazoliumtosylate (DAST) as the insulator layer and Silver (Ag) as the metal layer, in the simple MIM cavity [111] configuration. The total thickness of the structure is only 120 nm, where the thickness of layers (metal, insulator, and metal) is 25, 70, and 25 nm, from the top to the bottom. Description of optimization procedure along with conceptual analysis is presented. Moreover, a simple approach has been suggested for the determination of the resonance wavelength by the use of circuit theory, which has not yet been applied to the MIM structures, to the best of our knowledge. The passband region of the designed filter continuously covers the whole visible region by tuning the applied voltage, so that the output color changes from violet to red by changing the voltage from -12 to +12 volts. The device has the capa-

bility of functioning with negative voltages, which makes it relatively power-efficient. It can have applications in optical communication and switching, dynamic displays, holograms, imaging devices, light emitting structures, and smart color-tunable windows.

6.2 Calculation and Analysis

Figure 6.1(a) depicts the 3D schematic of the structure. It consists of two metal layers and an insulator layer sandwiched between them. Both metal layers should be thin so that the top metal layer allows the incident light couple into the structure, and, the bottom layer allows the light couple out of the structure and have a significant transmission. In other words, the MIM configuration acts like a Fabry-Perot cavity, in which the metal layers play the role of partially transparent mirrors and the dielectric layer fills the space between the mirrors. The physical phenomenon taking place in this cavity is that the incident light is partially reflected from the top metal-air interface, and partially penetrates into metal, and eventually penetrates into the cavity. The light is reflected back and forth between two semi-transparent metals and travels in the dielectric region of the MIM resonator. In each reflection, some part of light transmits through the bottom metal layer, eventually leading a significant portion of the incident light to be transmitted through the structure. Because of the Fabry-Perot-like behavior of the MIM cavity, not all the spectrum of the incident light gets trapped inside the cavity. Only spectral components corresponding to the resonance wavelength of the MIM cavity get trapped and eventually transmitted. The resonance wavelength of the MIM cavity, which determines the narrow transmission spectrum of this MIM filter, follows the equation [30]:

$$2 \left(\frac{2\pi}{\lambda_{res}} \right) n_i d_i + \varphi_b + \varphi_t = 2\pi m \quad (6.1)$$

where λ_{res} , n_i , and d_i denote the resonance wavelength of the MIM filter, the refractive index, and the thickness of the dielectric layer, respectively. The second and third terms in the left-hand side, φ_b and φ_t , denote the phase shift due to reflection from the bottom and the top metal layers, respectively. Finally, m in the right-hand side is an

integer that determines the order of the MIM cavity mode. For the light to be trapped inside the MIM cavity, the phase condition in equation (6.1) must be satisfied. This means that the phase accumulated in a full round-trip inside the cavity, which includes the effect of reflection from the metal layers (left-hand side of equation (6.1)) should be an integer multiple of 2π to enable the constructive interference of partially transmitted waves in each full round trip. Eventually, this constructive interference leads to the enhanced transmission of light through the cavity.

To calculate the transmission for the studied structures, the Transfer Matrix Method (TMM) is adopted in this work. The TMM calculations are verified by using Finite Difference Time Domain (FDTD) simulations performed by Lumerical FDTD solutions, a commercial FDTD software package [113]. Details of the TMM calculations are presented in the Supplementary Information.

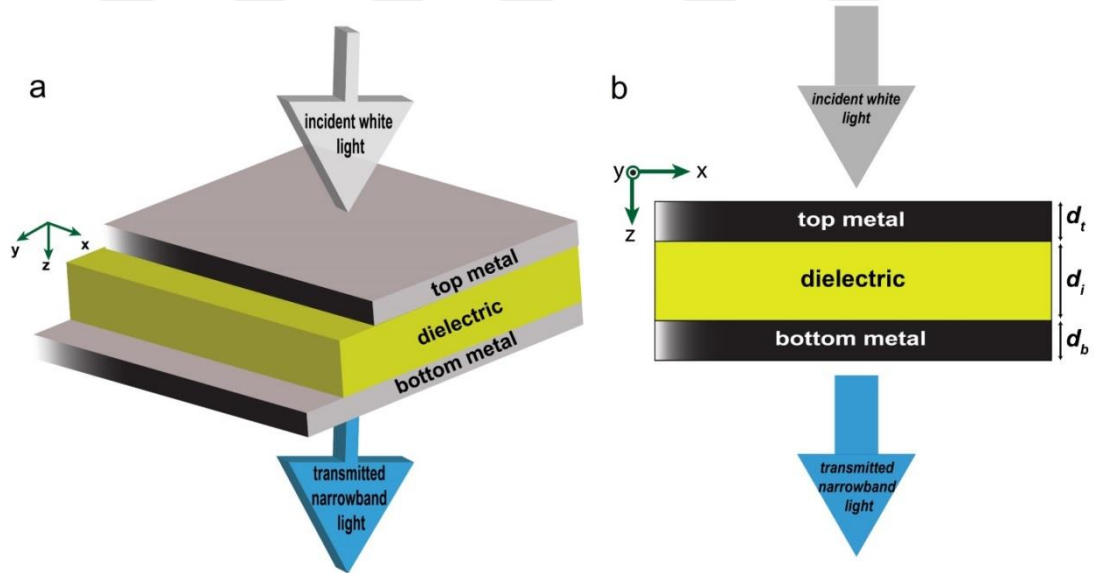


Figure 6.1: (a) 3D schematic of the MIM filter and, (b) the cross-section of the structure in (x,z)-plane. The names assigned to the thickness of layers are demonstrated in (b).

Now, we present the circuit model of the studied structure, which is built by using the transmission line (TL) theory. It is noteworthy that circuit models are quite universal. They have successfully been used in analysis and design of various optical [135], terahertz [64, 65, 136], and microwave [137] structures. In our case, the circuit

model significantly simplifies the calculation of the reflection phases for the bottom and the top metal layers, i.e., φ_b and φ_t . The rigorous electromagnetic approach is significantly more complex compared to the circuit theory approach we use here. It has to be mentioned that, to our knowledge, the circuit theory approach has not been applied in earlier works for the reflection phase calculations in MIM structures. In addition to the simplicity, it allows us to predict the resonance wavelength with perfect accuracy, as will be demonstrated in the next section.

The circuit model of the proposed structure is presented in Fig. 6.2. Z_t , k_t , and d_t are the normalized (to free space impedance) characteristic impedance, propagation constant, and the thickness of the transmission line for the top metal layer. By changing the subscripts of the same parameters from t to i or b , the similar parameters are introduced for the dielectric or the bottom metal layer, respectively. Z_0 is the free-space impedance and its normalized value is 1.

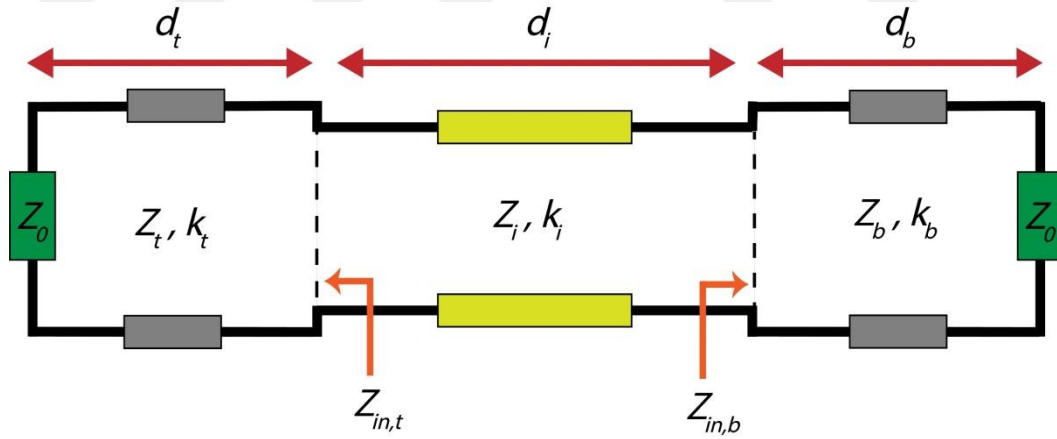


Figure 6.2: Circuit model of the structure using the transmission line theory. The incident light approaches from the left side.

In order to calculate the reflection coefficient of the top and bottom metal layers, one needs the input impedances seen from the metal-dielectric interfaces towards the metal layers. The input impedance seen from the interface of the top metal layer and the dielectric layer is denoted by $Z_{in,t}$ in the Fig. 6.2. $Z_{in,b}$ in this figure is the same parameter corresponding to the bottom metal layer. $Z_{in,t}$ is indeed the transformed impedance of air through the top metal layer. It is obtained using the following equation [112]:

$$Z_{in,t} = Z_t \frac{1 - jZ_t \tan(k_t d_t)}{Z_t - j \tan(k_t d_t)} \quad (6.2)$$

$Z_{in,b}$ can be calculated similarly, just by changing the subscripts from t to b in equation (6.2).

The reflection coefficients of the top metal layer and the bottom metal layer are named as r_t and r_b , respectively. r_t can be calculated as follows:

$$r_t = \frac{Z_{in,t} - Z_i}{Z_{in,t} + Z_i} \quad (6.3)$$

r_b is calculated similarly, by replacing $Z_{in,t}$ with $Z_{in,b}$. r_t and r_b are, in general case, complex numbers which can be written in the phasor form as follows:

$$r_t = |r_t| \angle \phi_t \quad (6.4a)$$

$$r_b = |r_b| \angle \phi_b \quad (6.4b)$$

Now, the calculation methods are elevated in detail, so we can calculate the transmission spectrum of an MIM filter. The top and the bottom metal layers both are chosen to be Ag with the thickness of 25 nm. The refractive index and the thickness of the dielectric layer are 2.2 and 70 nm, respectively. The thickness of the dielectric layer is chosen to have the resonance wavelength and the transmission band at the visible range. Fig. 6.3 shows the transmission spectrum of this MIM structure, which is calculated by using the TMM method.

To show that the resonance wavelength of the MIM cavity-based filter is correctly determined by (1), the total phase accumulated in a round-trip inside the cavity, ϕ_{tot} , which is given by the sum of the three terms in the left-hand side of (1), is calculated

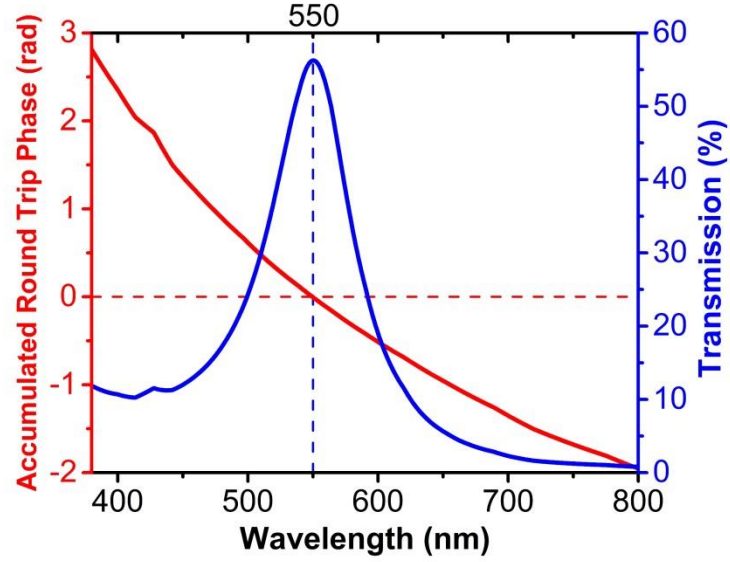


Figure 6.3: The transmission spectrum (blue line) of the MIM structure with the metal layers chosen as Ag and the values of n_i , d_t , d_i , and d_b being 2.2, 25 nm, 70 nm, and 25 nm, respectively; the corresponding vertical axis being on the right side of the plot in blue color. The accumulated round-trip phase (red line) according to equation (1) calculated by using the proposed circuit model; the corresponding vertical axis being on the left side of the plot in red color.

using the circuit model, as explained above. The obtained ϕ_{tot} of this structure is shown in Fig. 6.3 in red color together with the transmission spectrum shown in blue color. As follows from (1), ϕ_{tot} must be equal to an integer multiple of 2π , $m \geq 0$. In Fig. 6.3, we have shown the point, at which this condition is satisfied for $m=0$. As seen from Fig. 6.3, the resonance wavelength found from the phase condition perfectly matches the resonance wavelength or the peak wavelength of the transmission spectrum and is equal here to 550 nm.

For a better understanding of the electromagnetic phenomena occurring inside the structure, the magnitude of the electric (E) and magnetic (H) field distributions along the z-axis (normal to the layers of the structure), versus wavelength are shown in figures 6.4(a) and 6.4(b), respectively. The dashed vertical lines in these figures correspond to the resonance wavelength which is equal to 550 nm. From both figures, it is clearly seen that the structure supports a cavity resonance mode at this wavelength,

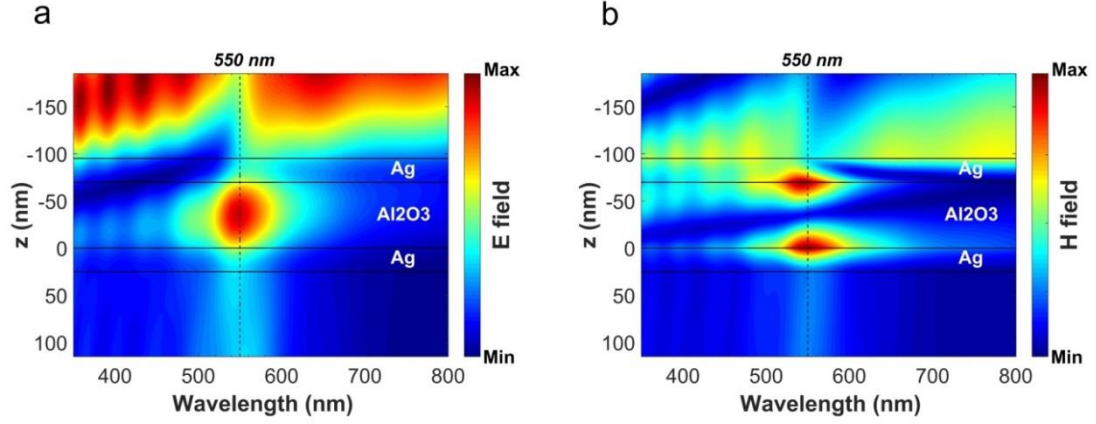


Figure 6.4: Contour plots of the magnitude of (a) the electric and, (b) the magnetic field in different parts of the structure, the transmission spectrum of which is shown in Fig. 6.3, versus the wavelength. The vertical dashed line crosses the wavelength of 550 nm, which corresponds to the resonance of this MIM filter.

which is trapped inside the dielectric layer. It becomes more obvious while looking at the E field distribution.

The other explanation of this phenomenon is that the field distributions convey the existence of the even mode supported by the MIM resonator at the resonance wavelength (see Fig. 6.4(b)). However, since the metal layers are not infinite in terms of their thickness, this supported mode is a leaky mode which can be coupled to the incident and transmitted plane waves leading to enhanced transmission at the resonance wavelength.

6.3 Optimization of the Structure

In this section, we will investigate the effect of the dimensions and materials on the transmission spectrum and optimize the proposed structure.

To explore the effect of the thickness of the dielectric layer on the transmission spectrum, we refer to equation (6.1). The term $n_i d_i$ on the left-hand side of this equation is actually the optical beam path inside the cavity. The main approach to tune the transmission spectrum is to modulate the beam path. It is obvious from the equation

that by increasing the path, the resonance wavelength or λ_{res} increases as well, or, in other words, it experiences a red-shift. Therefore, to tune the transmission spectrum, one can change either d_i or n_i . Figs. 6.5(a) and 6.5(c) show the transmission spectrum of the MIM structure for different thicknesses of the dielectric layer. The transmission profiles presented in Fig. 6.5(a) are calculated using the TMM method, and the ones shown in Fig. 6.5(c) are simulated by employing the Lumerical FDTD Solutions [113]. The metal layers are 25 nm thick and their material is chosen as Ag, and n_i is equal to 2.2. The red-shift in the transmission spectrum can be observed while increasing d_i . Figs. 6.5(b) and 6.5(d) demonstrate the transmission spectra of the structure for different values of n_i , which are calculated using the TMM and the FDTD methods, respectively. d_i is assumed to be 70 nm thick and the metal layers are completely the same as the ones for Fig. 6.5(a). It can be seen that by increasing n_i , the resonance wavelength of the transmission increases. To obtain a more precise image about the spectrum tuning effects, Figs. 6.5(e) and 6.5(f) depict the contour plots of the transmission spectrum versus the wavelength for varying values of d_i and n_i , respectively. Both contour plots are calculated using the TMM method. The corresponding color to each wavelength is also shown below the plots of Figs. 6.5(e) and 6.5(f) to help visualizing the change of the output color. It is seen from Fig. 6.5(f) that a less than twofold difference in n_i is required to cover the whole visible spectrum. However, the important restriction is that the above-mentioned tunings can be obtained when the changes are made at the stage of fabrication, i.e., by either changing the dielectric thickness or replacing the dielectric material with another dielectric with a different refractive index.

Here, by using an EO dielectric material, the refractive index of which changes while being under variations of applied voltage, the tunability of the transmission spectrum, like that in Fig. 6.5(b), can be realized in an active way after the fabrication. This is the basis of this work, which will be explored in the next section.

In order to design a color filter with a reasonably high quality factor (i.e., narrow bandwidth) and at the same time to have an appropriate transmission intensity, the choice of metals and the optimization of their thickness are of high importance. As it will be shown below, it is critical for the color filter performance.

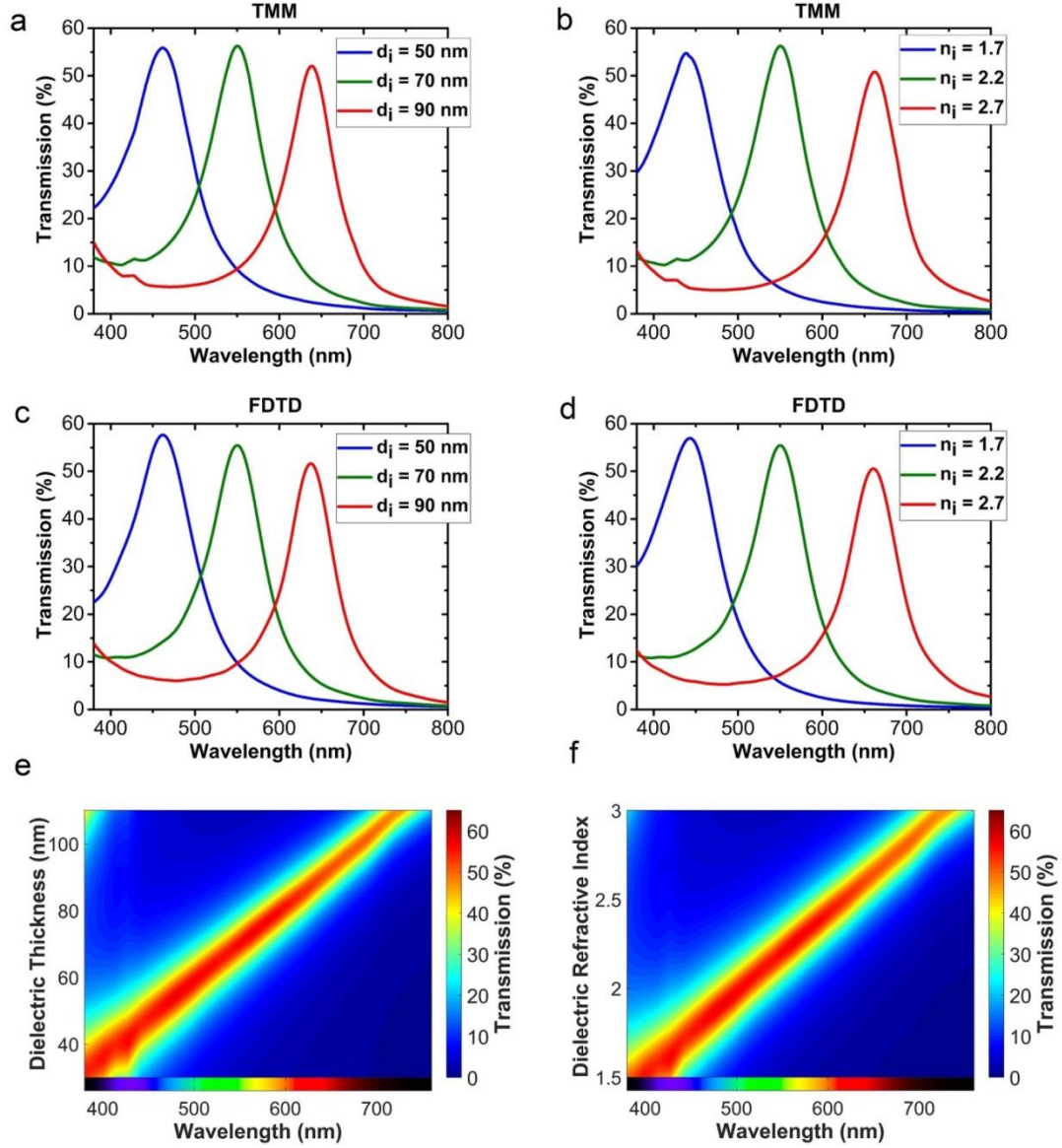


Figure 6.5: The transmission spectrum of the MIM filter calculated for different values of d_i by using (a) the TMM, and, (c) the FDTD method, and for different values of n_i by using (b) the TMM, and, (d) the FDTD method. Contour plots of the transmission spectra of the MIM filter (e) for varying values of d_i when n_i is 2.2, and, (f) for varying values of n_i when d_i is 70 nm, versus the wavelength, which are calculated by TMM, and, the output color as a function of wavelength is also shown below the plots in (e) and (f). In all cases, the material of both metal layers is chosen as Ag and the layers are 25 nm thick.

Figure 6.6(a) depicts the transmission spectrum of the MIM structure for n_i and d_i being equal to 2.2 and 70 nm, respectively, in the case of using all possible four com-

binations of Silver (Ag) and Gold (Au) as the top and the bottom metal layers with the thickness of 25 nm. The two metal names in the legend of the figure, left and right ones, correspond to the top and the bottom metal layers, respectively. It can be seen that the best result is obtained in the case of using Ag for both the top and the bottom metal layers, both in terms of the transmission strength and the higher quality factor. It can be observed that by replacing Au and Ag in the positions of the top and the bottom metal layers, the transmission spectrum remains the same. The complex refractive index data of Ag are taken from Palik [138] (0-2 μ m) and for Au, they are taken from Johnson and Christy [139].

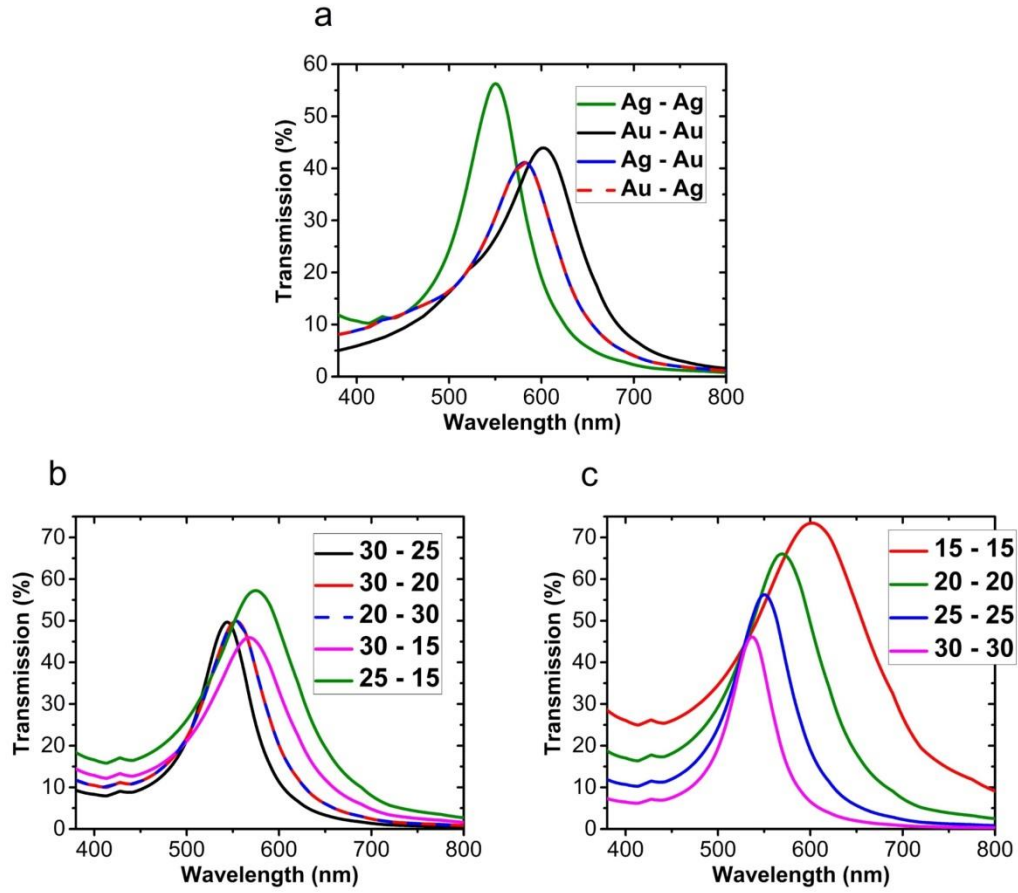


Figure 6.6: The transmission spectrum of the MIM filter calculated (a) for all possible combinations of Ag and Au used in the metal layers, and for different pairs of (b) unequal and (c) equal values of d_t and d_b when d_i is 70 nm and n_i is 2.2, and both the metal layers are chosen as 25 nm thick Ag.

Now, as the final step for the optimization of the structure, we investigate the effect of the thickness of the top and the bottom metal layers. Figure 6.6(b) shows the transmission spectrum for the case of $d_t \neq d_b$. The results also show that similar to replacing one metal by another, like in Fig. 6.6(a), there is no difference when interchanging the thicknesses of the top and the bottom metal layers. Fig. 6.6(c) demonstrates the calculated transmission of the structure when $d_t = d_b$. It is obvious from Figs. 6.6(b) and 6.6(c) that by choosing thinner metal layers, the transmission amplitude increases, however, this comes at the cost of losing the quality factor and the sharpness of the spectrum of the color filter. Taking this trade-off behavior into account, based on the calculations presented above, we choose the thickness of each of the top and the bottom metal layers to be 25 nm to achieve a reasonable compromise between the quality factor and transmission strength.

From Fig. 6.6 it is observed that by changing the thickness or the material of the metal layers, the resonance wavelength shifts as well. This may seem counter intuitive at first, but it can be clearly explained based on the previously presented results for the reflection phases, which are obtained from the circuit model. Indeed, any change in the choice of metal or the thickness of the top metal layer changes the values of Z_t , k_t , or d_t . Subsequently, the value of $Z_{in,t}$ (equation (6.2)) and therefore the values of r_t and especially φ_t (equation (6.3)) are changed. In a similar manner, any change in the bottom metal layer leads to a change in the value of φ_b . Therefore, the changes of the metal layers modulate the values of reflection phase shifts, and this leads to that the resonance wavelength experiences a shift (see equation (6.1)).

To briefly summarize, we have so far optimized the color filter by choosing the appropriate metal to be Ag and the thickness of the top and bottom metal layers to be 25 nm, both. We also explained in-detail the effects exerted by the basic parameters on the quality factor, transmission intensity, and the resonance wavelength of this MIM filter.

6.4 Electrical Tuning

As shown in the previous section, a less than twofold difference in n_i should be sufficient to cover the whole visible spectrum. The required difference can be realized by implementing the electrical tunability through the use of DAST, an EO material, as the dielectric layer. DAST has a large EO coefficient equal to 3.41 nm/V [97, 140-142]. The refractive index of DAST is a function of the applied voltage and can be written as

$$n_i = n_0 + \frac{dn}{du} \left(\frac{V}{d_i} \right) \quad (6.5)$$

where d_i denotes the thickness of DAST sandwiched between the two metal layers and V is the applied voltage; $dn/du = 3.41$ nm/V is the EO coefficient with u being the applied electric field, and $n_0 = 2.2$ is the refractive index of DAST at $V=0$.

Fig. 6.7(a) depicts the schematic of the structure with applied voltage. Fig. 6.7(b) shows the transmission spectrum for different values of the applied voltage when d_i is 70 nm. It is obvious from this figure that by varying the voltage from -12 to +12 volts almost the whole visible spectrum can be covered by the output transmission color of this MIM filter. Fig. 6.7(c) demonstrates the continuous tuning of the transmission spectrum by plotting the contour plots of the transmission versus the wavelength and applied voltage. For a better visualization of the color tunability, the output color as a function of wavelength is also shown below the plot of Fig. 6.7(c).

The 3D surface plot of the accumulated phase in a round-trip inside the cavity, φ_{tot} , is shown in Fig. 6.7(d) as a function of wavelength and the applied voltage. The intersection of the surface corresponding to φ_{tot} with the plane of zero-phase (the first integer multiple of 2π) determines the points of resonance and, thus, the expected peaks of transmission spectra. It can be seen that the intersection with zero-phase plane (the resonance wavelength) increases from around 400 nm to around 700 nm by increasing the applied voltage. The resonance wavelength versus the voltage is plotted in Fig. 6.7(e). Here, the wavelengths are calculated by finding the peak of the transmission spectrum from the TMM method, and are also calculated from the intersection of

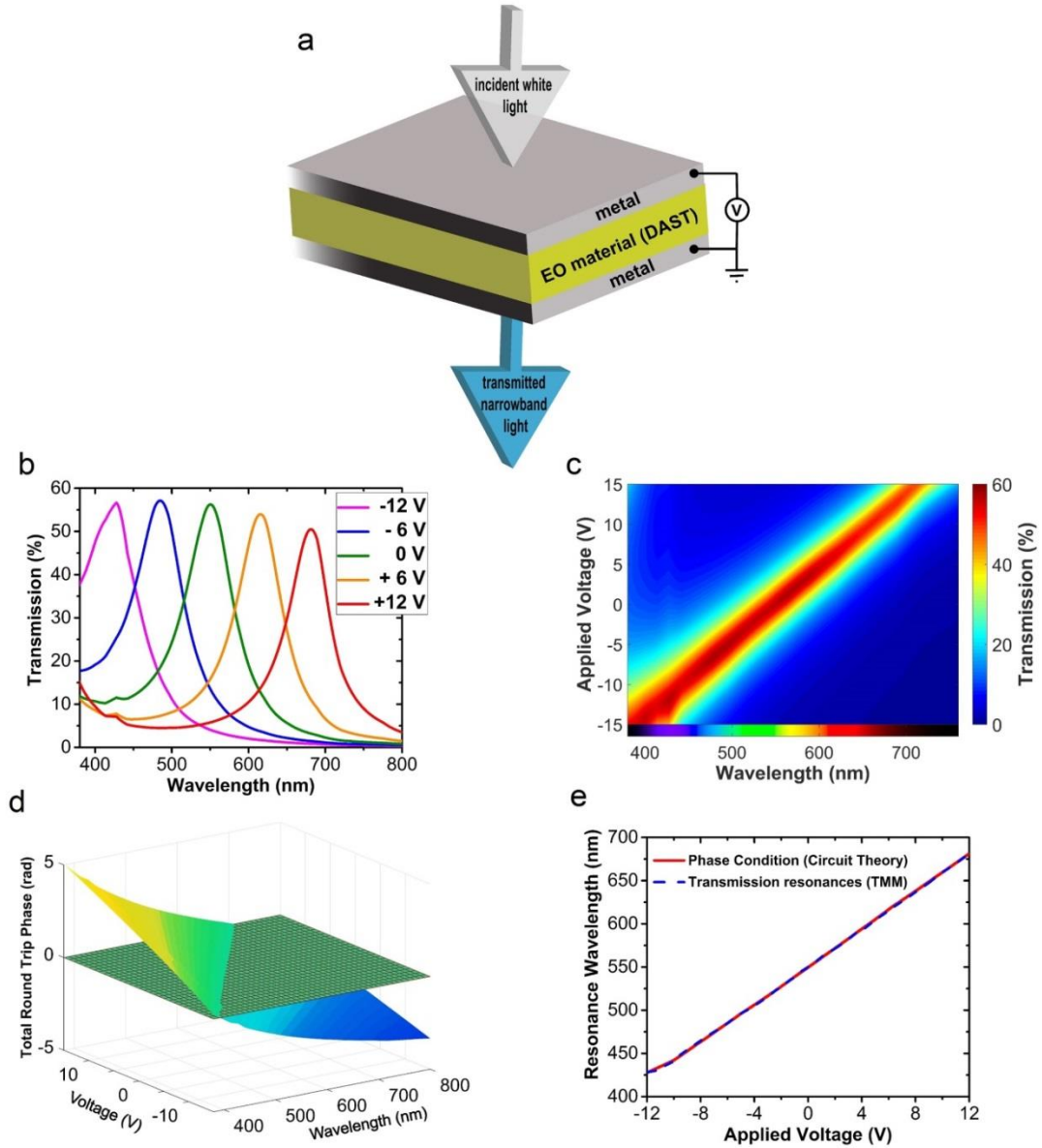


Figure 6.7: (a) 3D schematic of the electrically tunable color filter, (b) the transmission spectrum of the color filter for different values of the applied voltage, (c) contour plots of the transmission spectra at continuous variation of applied voltage, the output color as a function of wavelength is also shown below the plot, (d) 3D surface plot of ϕ_{tot} versus wavelength and applied voltage; the meshed plane shows the zero-phase plane, and, (e) the resonance wavelengths versus the applied voltage calculated from the phase condition (equation 6.1) and from the TMM method.

ϕ_{tot} with zero-phase plane shown in Fig. 6.7(d). There is a perfect agreement between the two methods. The results presented in Fig. 6.7(e) clearly show that the fine and

continuous tuning of the output color can be achieved by varying the applied voltage from -12 V to +12 V. Based on these results, the average shift in the resonance wavelength for every 10 V increment in applied voltage is 105.4nm, which is a great number compared to the previous works [68, 97, 143, 144]. For instance, in the design proposed in [59], a less than 50 nm shift of the resonance wavelength is obtained in the visible regime by the application of 160 V. In the design proposed in [17], a 40 nm shift is obtained with 50 V. Also in [46], the resonance peak shifts 22.5 nm per 10 V. Finally, in [4], a less than 100 nm shift is achieved in the near-infrared region by a 70 V applied voltage. Therefore, our proposed design is really power-efficient.

It is also expected that the suggested structure has the potential of electrical tunability in the case of oblique incidence. By applying the necessary changes to the

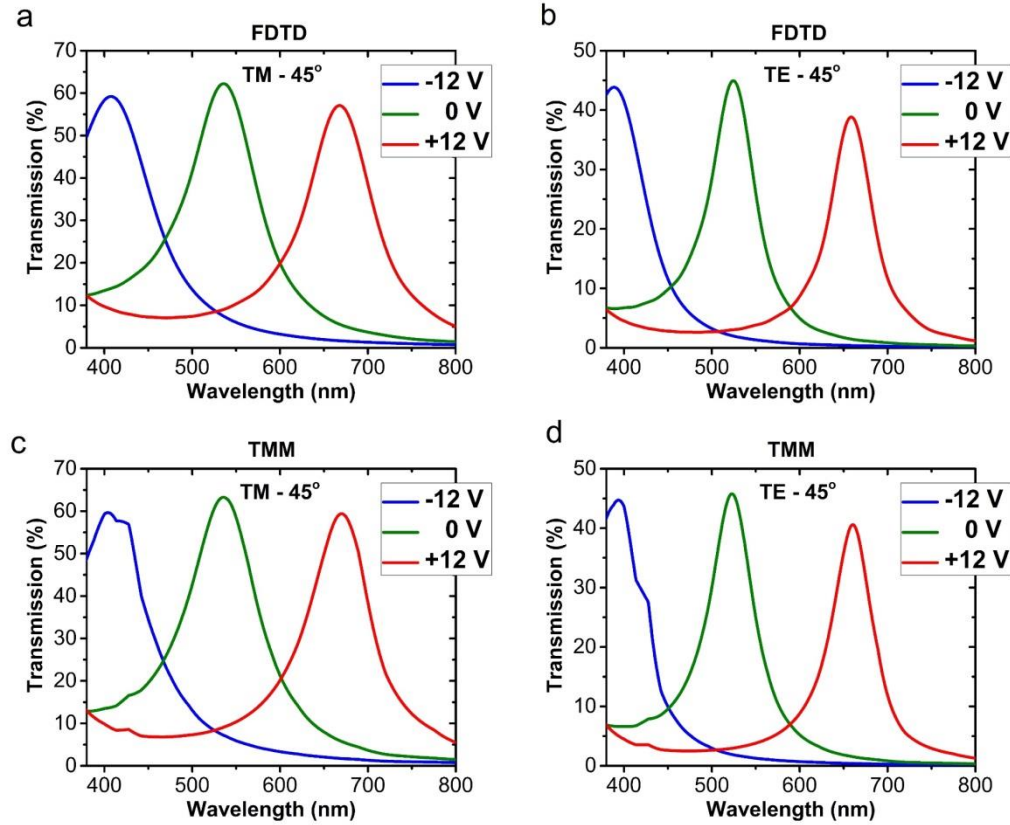


Figure 6.8: The transmission spectrum of the electrically tunable color filter for different values of the applied voltage at the incidence angle of 45 degrees, for (a) TM and (b) TE polarization, calculated by the FDTD method, and for (c) TM and (d) TE polarization, calculated by the TMM method.

TMM calculations, the transmission spectrum of the structure for the incidence angle of 45 degrees has been calculated. The results are shown in Fig. 6.8 for both TM and TE polarizations. They confirm that the tunability is nearly the same as at normal incidence; i.e., almost all the visible range gets covered by sweeping the voltage between -12 and +12 volts.

The obtained results show that by the use of this simple geometry, and by the correct and optimized choice of materials and dimensions, a powerful tuning of the color of the MIM cavity based filter in the visible region can be obtained. The proposed structure covers the whole visible region by varying voltage only from -12 to +12 volts, so it is also power-efficient. This structure does not require any patterning and lithography and has a total thickness of 120 nm, i.e., it is ultra-thin and significantly light. Therefore, it is perspective for large-area and mass production of electrically tunable color filters.

6.5 Conclusion

The general concept, theoretical background, and in-detail design steps of an MIM cavity based, electrically tunable color filter are presented. The effect of each design parameter has been investigated. An unprecedentedly simple approach has been suggested for the precise calculation of the resonance wavelength of the MIM structures, which is based on the circuit theory. The optimized electrically tunable structure which has the total thickness of 120 nm gets tuned from transmission output spectrum of the violet color to the red color, by only varying the applied voltage from -12 to +12 volts. The electrical tuning is continuous and covers the whole visible region. The proposed design has an important advantage of being lithography-free, which makes it a great candidate for large-area and mass production of tunable color filters. Our design is also relatively power-efficient compared to the earlier works. The presented structure can be used in a large number of applications such as optical communication and switching, displays, imaging devices, and color-tunable windows.

Chapter 7

Multifunctional Polarization-Sensitive Beam Deflector and Angular Filter

7.1 Introduction

In this section, we experimentally report a simple and cost-effective structure with an unprecedentedly wideband, wide-angle, and highly efficient beam deflection to the -1st reflection order in the Near-Infrared region. In addition, it has a strong switching efficiency between the TE and TM polarization for this behavior. Moreover, it shows multifunctional SF behavior for different diffraction orders and different polarizations at specific frequency domains. In particular, it performs as band-pass, low-pass, and band-stop spatial filter in different scenarios. In addition to these functionalities, the structure works as an efficient beam-splitter in for TE polarization at an specific wavelength range. The structure consists of silicon nanorods patterned on top of a dielectric spacer deposited over an Aluminum back reflector. Its simple design, along with the wideband, wide-angle, and efficient deflection, being polarization switchable, and having multi-functional SF and beam-splitting performances makes this structure a promising candidate for variety of applications including optical communications, sensing, detection, laser optics, spectroscopy, and imaging.

7.2 Results and Discussion

Three-dimensional and two-dimensional schematics of the structure are shown in Fig. 7.1(a) and (c), respectively. Incident, 0 order (specular reflection) and -1 order reflection beams are also shown in Fig. 7.1(c). As it can be observed, the structure consists of a periodic array of Si nanorods patterned on top of an SiO₂ (silica) spacer layer coated over an Aluminum (Al) layer. As shown in Fig. 7.1(c), p , w , h , d , and b denote the period, width of nanorods, height of nanorods, thickness of silica layer, and thickness of the bottom Al layer, respectively. In order to block all the transmission, the bottom metallic layer must be sufficiently thick therefore we have conservatively chosen b to be 150 nm. Moreover, the optimized values of p , w , h , and d are 1250, 150, 180, and 150 nm, respectively. It is noteworthy that the dimensions are not strictly chosen and simulations have confirmed that the structure has a good fabrication tolerance. In other words, the result is almost robust to small deviations of the dimensions from the abovementioned values. Moreover, even big deviations from the optimal dimensions may affect the desired result of the structure, but still keeps the main performance features such as high efficiency and wideband behavior. Therefore, the wideband and wide-angle functionality of the structure is not an accidental coincidence and the result of optimizing the dimensions, and is rather the result of the nature of this design. Moreover, Fig. 7.1(b) demonstrates Scanning Electron Microscopy (SEM) image of the top view of the fabricated sample. For more information on the sample fabrication, see the “fabrication” section.

The deflection efficiency is defined as the ratio of the power transferred to any specific reflection order to the total reflection, where the total reflection is the sum of the powers transferred to all reflection orders. The deflection efficiency (D.E) of the i 'th order is calculated as follows:

$$D.E = \frac{R_i}{R_{tot}} = \frac{R_i}{\sum R_i} \quad (7.1)$$

where R_i and R_{tot} denote the reflection of the i 'th order and the total reflection, respectively.

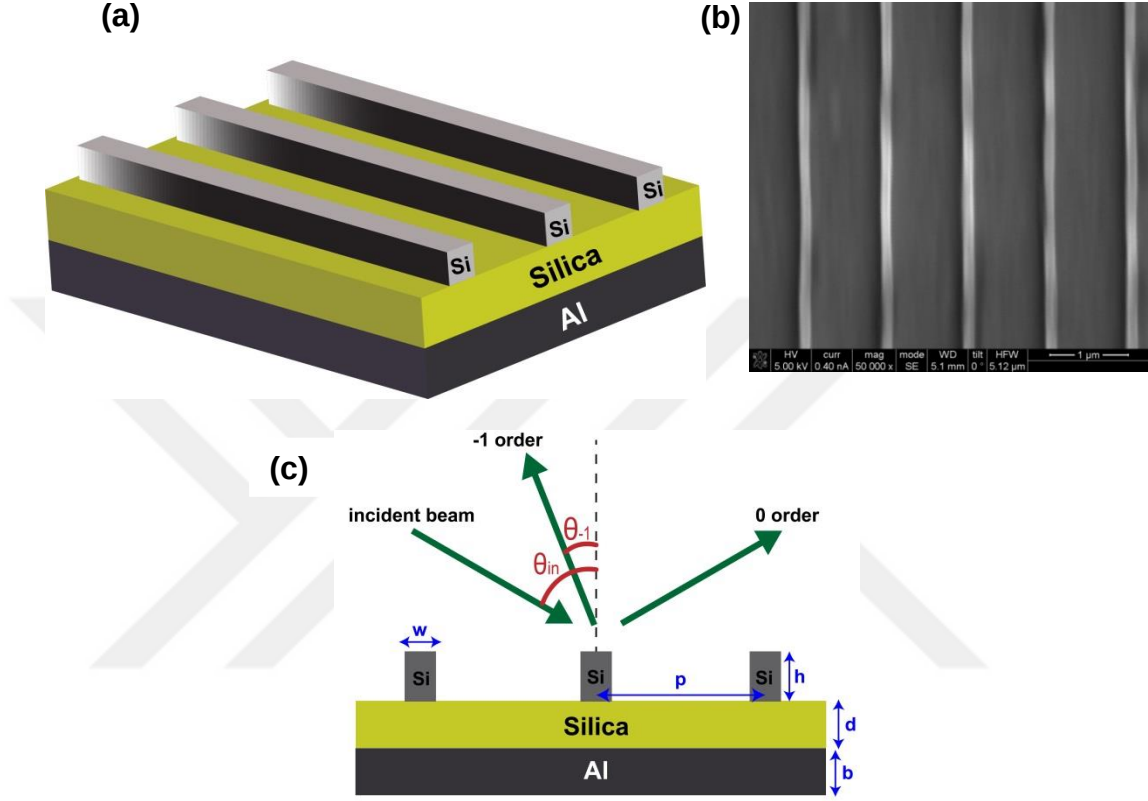


Figure 7.1: a) 3D schematic of the structure, b) SEM image of the top view of the fabricated sample, and c) 2D cross-section of the of the structure along with the schematic of the incident, 0 order reflection, and -1 order diffraction beams.

The structure is designed in a way that it has a very efficient wide-angle and wide-band deflection to the -1 order for the TE polarization (when the electric field of the incidence beam is parallel to nanorods). This feature is polarization sensitive and can be strongly switched by changing the polarization, i.e. the structure has almost zero deflection efficiency in -1 order for the TM polarization (when the electric field of the incidence beam is perpendicular to nanorods).

To demonstrate this issue, the simulated deflection efficiency of the -1 order (D.E (-1)) of the structure as a function of incidence angle is plotted and shown in Fig. 7.2(a), at four different constant wavelengths, for the TE polarization. The simulations are carried out using FDTD Lumerical software [113]. For more information on

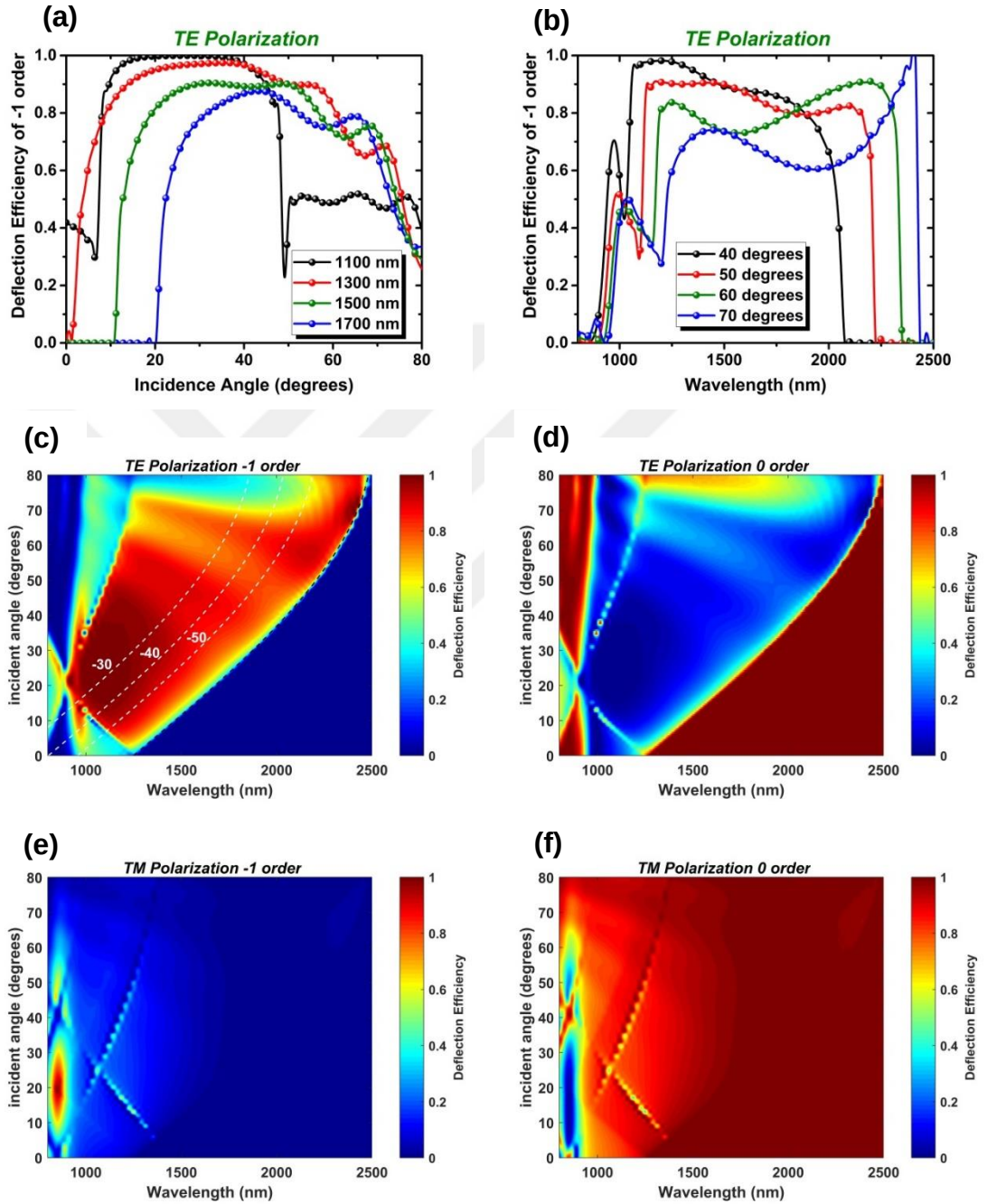


Figure 7.2: Simulated deflection efficiency of -1 order of the structure for TE polarization as a function of a) incidence angle and b) wavelength. Measured deflection efficiency contour plots of a) TE polarization -1 order, b) TE polarization 0 order, c) TM polarization -1 order, and d) TM polarization 0 order, as a function of wavelength and incidence angle. The white dashed lines in (c) represent iso-lines of constant -1 order reflection angles of -30, -40, and -50 degrees. The black dashed line in (c) represent the border where -1 order starts to appear.

the simulation details, see the “simulation” section. The four wavelengths are chosen as 1100, 1300, 1500, and 1700 nm. It can be observed that at a constant wavelength, the structure has high D.E (-1) in a wide angle range for the TE polarization.

Now, in order to observe the wideband behavior of the structure in the frequency (wavelength) domain, the simulated D.E (-1) of the structure is shown as a function of wavelength for four different incidence angles in Fig. 7.2(b) for the case of TE polarization. Similarly, it can be observed that the structure shows a very strong D.E (-1) in a wide wavelength range and for various incidence angles, for the case of TE polarization.

It was already demonstrated that the structure not only has a wideband and wide angle highly efficient deflection to the -1st order. Therefore, it is better to discuss D.E (-1) as a function of both angle of incidence and wavelength and plot it for both polarizations. In order to calculate the deflection efficiency of any specific reflection order, we must have the total reflection which is distributed among all the reflection orders (see Eq. (7.1)), so that we have calculated the reflection power of all reflection orders. D.Es of -1st and 0th orders of reflection are shown as contour plots as a function of wavelength and incidence angle for the TE polarization in Figs. 7.2(c),(d), respectively, and for the TM polarizations in Figs. 7.2(e),(f), respectively.

It can be observed in Fig. 7.2(c) that for TE polarization, the structure retains high deflection efficiency for -1 order in an ultra-wideband and wide angle range, generally in NIR. On the other hand, as can be observed in Fig. 7.2(e) that, for the case of TM polarization, the structure shows very low D.E (-1) in the same range of wavelength and incidence angles. Moreover, it can be observed in Fig. 7.2(d) that the deflection efficiency of the 0th order (specular reflection) of the TE polarization is very low in the same region where D.E (-1) of this polarization is high. In other words, in a wideband and wide-angle range, the reflection power is transferred to the -1 order from the specular (0) reflection order, for the case of TE polarization. It can be observed in the right edge of efficient deflection region that this power transition from the 0 to the -1 order is very sharp and strong, and the reflected power coupled to the 0 order immediately drops significantly by entering the deflection region. This edge is showed by black dashed line in Fig. 7.2(c) which represents the border at which -1

order of reflection starts to appear. Furthermore, it can be seen that D.E (0) is high as expected, for the TM polarization. Therefore, this phenomenon can be interpreted in another way. By switching the polarization from TE to TM, the reflected power switches from zero to -1 reflection order, for a wide range of incidence angles and for a broad wavelength range.

Figure 7.3 shows the experimental results measured from the fabricated sample, the SEM image of which is shown in Fig. 7.1(b). Figures 7.3(a) and (b) demonstrate contour plots of D.E (-1) as a function of wavelength and incidence angle, representing measured and experimental results, respectively. It is observable from the measured range that there is a good agreement between the experiment and simulation and in addition, the fabricated device also shows the expected broadband and wide-angle efficient deflection in the measured range. Moreover, Fig. 7.3(c) demonstrates the measured D.E (-1) as a function of wavelength for different incidence angles ranging from 55 to 75 degrees, for the TE polarization. Also, Fig. 7.3(c) illustrates the measured D.E (-1) as a function of incidence angle for fixed different wavelength values, for the same polarization.

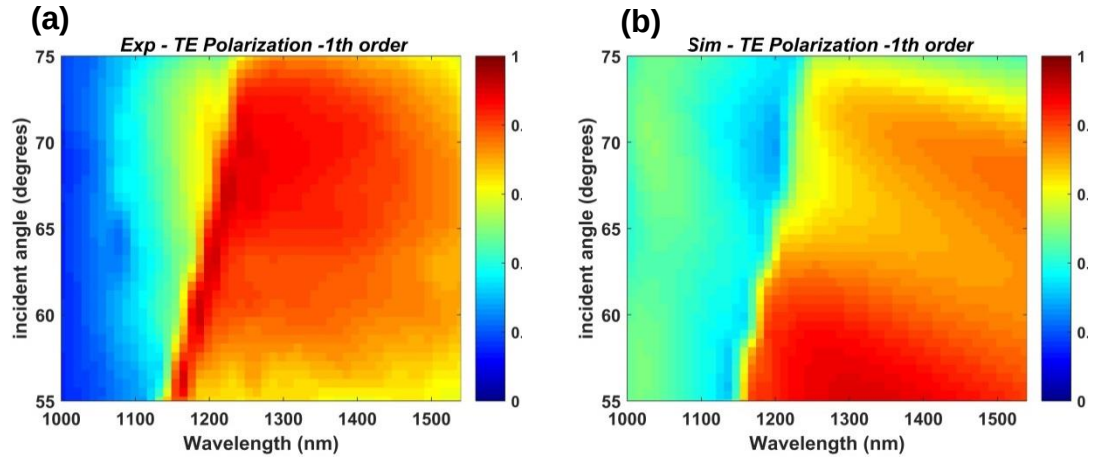


Figure 7.3: Measured deflection efficiency contour plots of -1 reflection order of the structure for a) TE and b) TM polarizations, as a function of wavelength and incidence angle.

As it can be observed in the plots of Fig. 7.3, the wavelength range and incidence angle of the measurements of D.E (-1) are limited to 1000-1540 nm, and 55-75 degrees, respectively. This is due to the fact that the detector had to be in a specific po-

sition for each wavelength and for each incidence angle to measure an specific order of the reflected beam, and the measuring device (J.A. Woollam Co. Inc. VASE Ellipsometer) has limitations in terms of the position of detector with respect to the source. For more information on the measurement details, see the “characterization” section.

The angle of the output-coupling i 'th order reflected wave is a function of both wavelength and incidence angle, and is calculated as follows:

$$\sin(\theta_{out,i}) = \sin(\theta_{in}) + \frac{i\lambda}{p} \quad (7.2)$$

where p , λ , θ_{in} , and $\theta_{out,i}$ denote the period, wavelength, incidence angle, and i 'th order reflection angle, respectively. Given the limitations of the rotation and position of the detector in the measuring device, for some wavelengths at some specific incidence angles, the -1 reflection order was not measurable.

For the purpose of a clear demonstration of variation of $\theta_{out,i}$ by θ_{in} and λ , iso-lines of constant output angle at -1 order are presented using white dashed lines for three different constant values of $\theta_{out,-1}$ of -30, -40, and -50 degrees as a function of wavelength and incidence angle.

In addition to the most important and main feature of this structure which is having a highly-efficient, wideband and wide-angle domain of deflection to the -1 order, and being polarization switchable for this behavior, it demonstrates multiple interesting functionalities as different types of spatial filter (incidence angle domain). First of all, around the wavelength of 900 nm, a band-stop spatial filter (SF) can be observed for the 0 order of TE polarization, which is shown in Fig. 7.2(d). To demonstrate it more clearly, Fig. 7.4(a) shows D.E (0) of the TE polarization as a function of incidence angle at three slices of fixed wavelengths of 890, 900, and 910 nm. A clear band-stop SF can be observed for these wavelength values at 0 order. Secondly, at wavelengths higher than around 1300 nm, we can observe a low-pass SF behavior in the 0 order of TE polarization in Fig. 7.2(d). To illustrate this issue, D.E (0) of TE polarization as a function of incidence angle at four slices of fixed wavelengths of 1500, 1700, 1900,

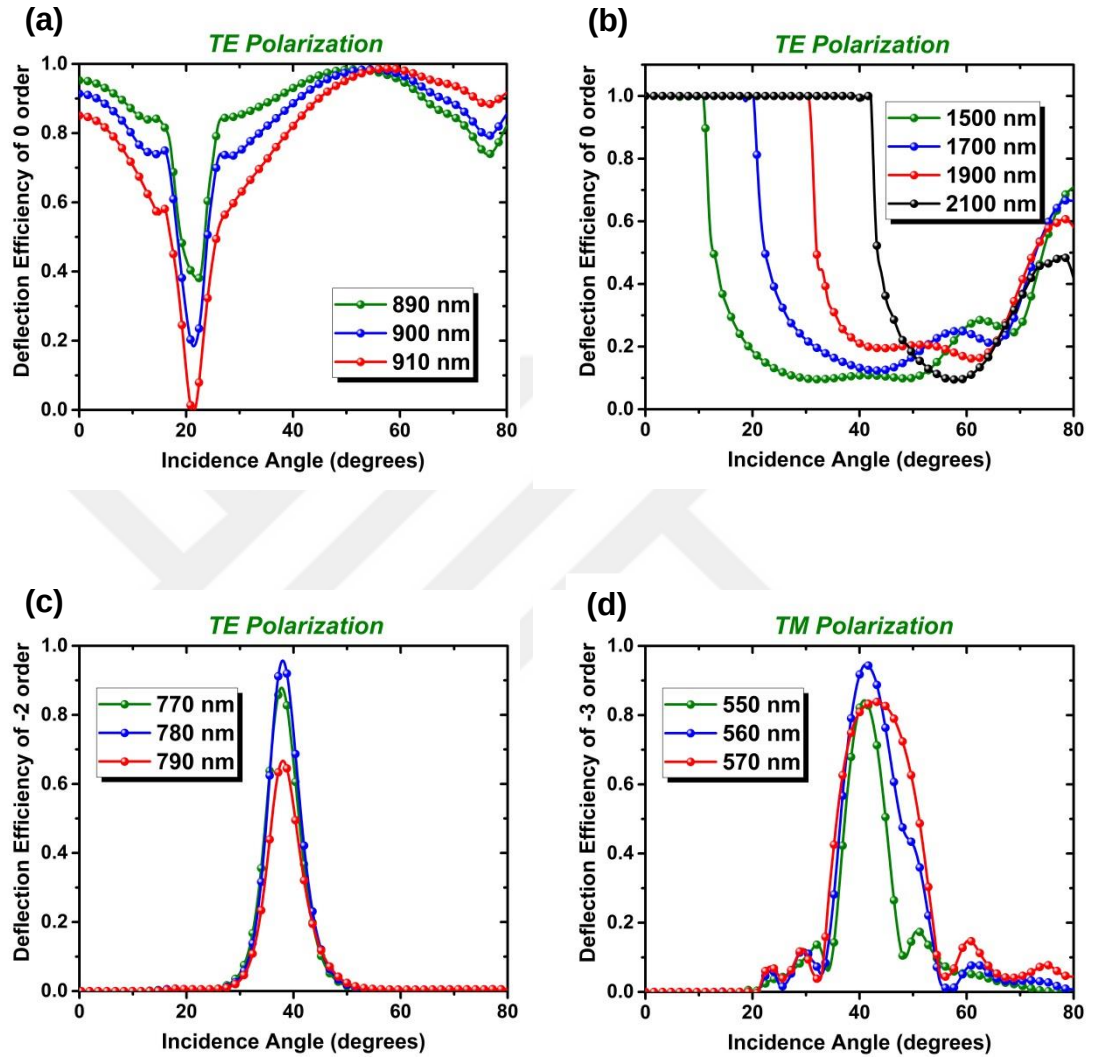


Figure 7.4: Spatial filter functionalities of the structure. Deflection efficiency of a) TE polarization at 0 order at the wavelengths of 890, 900, and 910 nm, b) TE polarization at 0 order at the wavelengths of 1500, 1700, 1900, and 2100 nm, c) TE polarization at -2 order at the wavelengths of 770, 780, and 790 nm, and d) TM polarization at -3 order at the wavelengths of 550, 560, and 570 nm.

and 2100 nm is plotted and shown in Fig. 7.4(b), where the low-pass SF behavior is available. As the wavelength increases, the right edge of the pass-band SF shifts toward higher angles which is also evident from the contour plot of D.E (0) in Fig. 7.2(d). Moreover, Fig. 7.4(c) demonstrates D.E (-2) of TE polarization as a function of incidence angle at three slices of fixed wavelengths of 770, 780, and 790 nm. Similarly, Fig. 7.4(d) depicts D.E (-3) of TM polarization as a function of incidence angle

at three slices of fixed wavelengths of 550, 560, and 570 nm. A band-pass SF behavior of the structure is quite obvious in both figures.

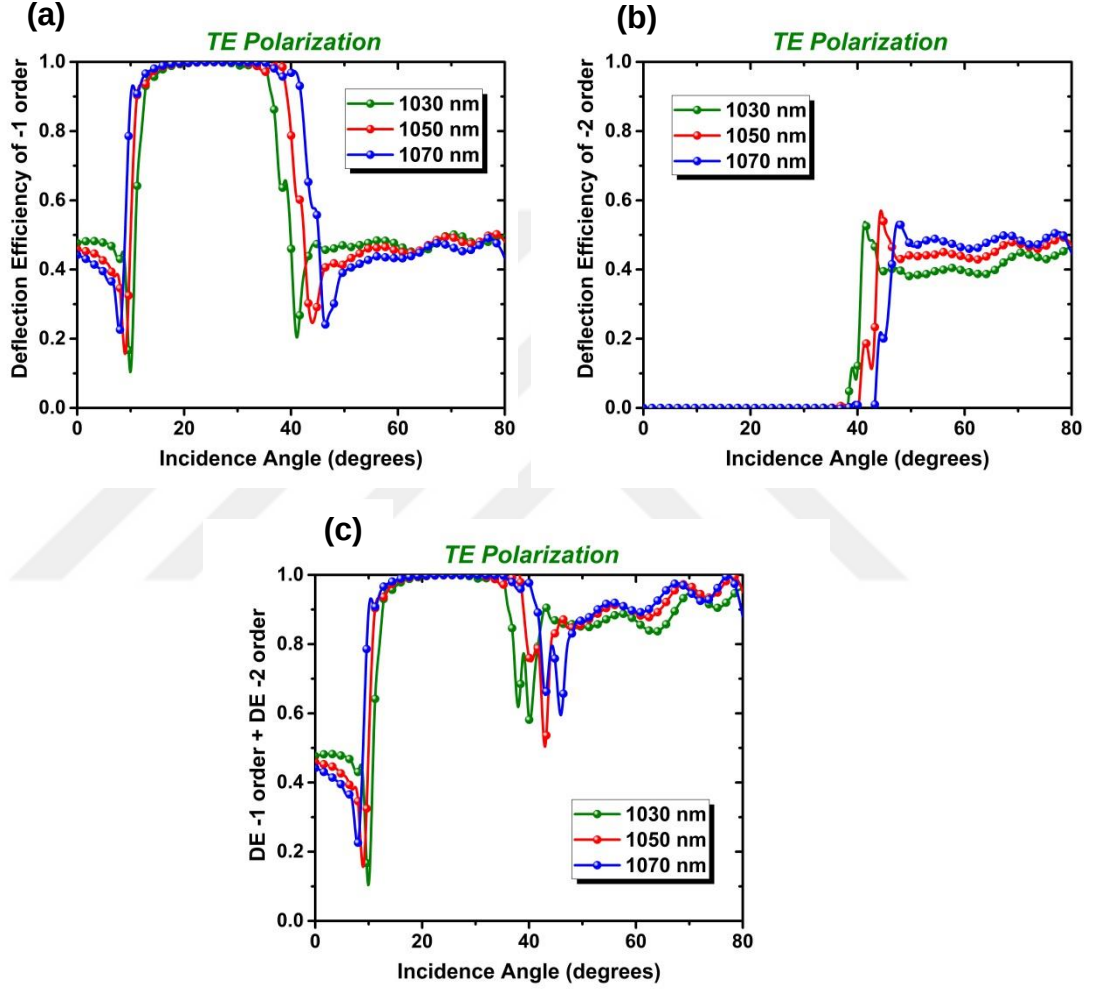


Figure 7.5: Beam splitter functionality of the structure. Deflection efficiency of TE polarization at a) -1 order, b) -2 order, and c) sum of -1 and -2 orders, at the wavelengths of 1030, 1050, and 1070 nm.

So far, it is shown that the structure, in addition to polarization switching feature and ultra-wideband and wide-angle beam deflection, functions as band-pass, band-stop, or low-pass spatial filter, depending on the reflection order and polarization. It was also shown that the structure partially shows polarization independent behavior for 0 order. In addition to these features, the structure shows beam-splitting functionality in a relatively wide-angle range for TE polarization. Figs. 7.5(a,b,c) demonstrate

D.E (-1), D.E (-2), and D.E (-1)+ D.E (-2), respectively, for TE polarization as a function of incidence angle at three slices of fixed wavelengths of 1030, 1050, and 1070 nm. It can be observed in these figures that for incidence angles approximately larger than 40 degrees, we have wide-angle beam-splitting between -1 and -2 orders of TE polarization around the wavelength of 1050 nm. It is evident from Fig. 7.5(c) that the sum of these two orders is close to unity and it can be observed in Fig. 7.5(a,b) that their sum is almost equally distributed among these two orders. In other words, the reflected beam power is almost split between these two reflection orders for a wide range of incidence angles.

7.3 Conclusion

A multi-functional, highly efficient, ultra-wideband and wide-angle beam deflector and spatial filter was experimentally presented. The structure consists of simple-to-fabricate elements. This design transfers the reflected power to -1 order in a wideband and wide-angle range in a very efficient way for the TE polarization. By switching the polarization from TE to TM, the structure does not show this deflective behavior. Therefore, it has polarization switchability. Moreover, it demonstrates multiple spatial filter functionalities including band-pass, band-stop, and low-pass angular filtering, depending on the polarization, wavelength, and reflection order. Furthermore, it shows a wide-angle beam splitting between the -1 and -2 order of TE polarization. Having high efficiency along with multiple functionalities which is accompanied by a simple design, this structure can serve as a promising candidate for optical communication, spectroscopy, sensing, and laser optic applications.

7.4 Methods

7.4.1 Simulation

The simulations are carried out using the commercial Lumerical FDTD software [113]. A 2D simulation environment is used in the xy coordinate system. periodic boundary condition is used along x-direction, and perfect electric conductor (PEC) boundary condition is used along y-axis on both sides. BFAST plane-wave is used as the source of the simulation and the source illuminates the structure in $-y$ direction. Above the source, there is a DFT monitor to collect the reflected beam. A modified scripting code taken from the software's website is used to calculate the power transferred to different reflection orders.

7.4.2 Fabrication

A 1 cm^2 piece of commercial Silicon wafer is used as the substrate. The substrate is cleaned using Acetone, Isopropanol, and water. Then, a 150 nm thick Al layer is coated over it with the rate of 0.5 Angstrom/s using a VAKSIS thermal evaporator device in the chamber pressure of $3\text{e-}6$ to $5\text{e-}6$ Torr. Afterwards, a 150 nm Silica layer is coated over the Al layer using VAKSIS electron-beam evaporator in the same rate and chamber pressure. After that, 180 nm Silicon is coated over it using the same e-beam evaporator device. Then, an area of 1 mm^2 is patterned using Electron Beam Lithography. Finally, the sample is etched using the lift-off process.

7.4.3 Characterization

Measurements are carried out using J.A. Woollam Co. Inc. VASE Ellipsometer. Two lenses are added and aligned on the source and detector side for aligning the beam on the patterned area over the sample. The measurement range of the detector starts from visible and ends in 1540 nm. Moreover, the angle between the source and detector

should at least be 30 degrees. The detector arm has the possibility of sweeping the detection angle with the desired step sizes.



Chapter 8

Conclusion

The thesis demonstrated and discussed three major functionalities of metamaterials. More specifically, broadband absorbers, electrically tunable color filters, and metasurfaces as beam deflectors and multifunctional angular filters. Most of the works carried out throughout the thesis were lithography-free which would make them a promising candidate for mass production.

For the first time, we used the metal of Manganese for broadband absorber applications and showed in-detail what a promising metal it is for such applications. It was demonstrated that this metal has better functionality compared to the best other candidates for broadband absorption and it is due to its appropriate optical properties. It was shown that the absolute value of the real part of permittivity of Mn remains very small compared to the other metals which leads to high field penetration inside it. Being a lossy metal, it absorbs the highly penetrated electromagnetic field, and as a conclusion, it makes a promising absorber metal. It was also shown that even the absorption of a single planar layer of Mn is significantly better than other metals. These properties were fully exploited in four different structures as broadband absorbers, three of which were lithography-free, and therefore, very promising for mass production. Such absorbers can be used in thermal emission, shielding, photovoltaics and energy applications, photodetectors, and thermal imaging. As a conclusion, we propose that Mn should be tried in almost all broadband absorbers in the design step. In addition, it would be nice to try the usage of the structure in chapter 5 as a solar cell

or water splitter and by having high and broadband absorption, we expect to see high conversion efficiency.

Moreover, it was shown that by the proper use of an electro-optic material abbreviated as “DAST” in the MIM configuration, lithography-free, electrically tunable color filter is achievable. By putting such a structure in the path of the white light beam and looking at it from the other side and ranging the applied voltage from -12 to +12 volts, we can see all the colors in the visible range. This structure can have nice applications in optical communications and displays. It would be beneficial to fabricate this structure and see the experimental realization of this design in a display configuration while there is no lithography needed.

While achieving efficient beam deflection is a challenge, obtaining it in a wide-angle and wideband range is more of a challenge. We showed that this challenge can be overcome by the proper design of the dimensions in Silicon nanorods. Moreover, at a time when many previous works use a whole new structure to obtain one angular filtering functionality, the same structure could serve as band-pass, band-stop, and low-pass angular filters, demonstrating multi-functional angular filtering functionalities. Such a structure would be very beneficial for optical communications, sensing, and imaging. As a future outlook, this work can be shifted to the visible region with the appropriate rescaling to obtain multi-functional angular filtering or deflection in the visible range and for different colors.

In the first chapter, the background and the thesis outline and classification was presented.

Chapters 2 to 5 were all based on broadband absorbers that exploited Mn. Compared to the simplicity of the structures, and considering the fact that the structures are lithography-free, the results were extraordinarily promising. For instance, in chapter 5, UV to FIR absorption was achieved by coating a single layer of Mn over a high-roughness substrate that includes pyramidal features on it. Such a result was not previously achieved even with the inclusion of lithography. Circuit modelling and Transfer Matrix Method calculations were presented in detail.

Chapter 6 presented the design of a lithography-free electrically tunable color filter that would cover the whole visible spectrum. As mentioned above, idea was to implement an electro-optic dielectric in the MIM configuration. The achieved results confirmed that -12 to 12 volts would be enough to cover the visible spectrum which makes the structure very power-efficient. Circuit model was also used here to calculate the reflection phases for the resonance mode of the cavity.

Finally, Chapter 7 demonstrated a relatively simple metasurface consisting of Silicon nanorods that would result in highly-efficient, wideband and wide-angle beam deflection. Moreover, the same structure showed multiple functionalities as different types of spatial (angular) filters.

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Appendix A

Publications

1. **Majid Aalizadeh**, A. Khavasi, B. Butun, and E. Ozbay, “Large-Area, Cost-Effective, Ultra-Broadband Perfect Absorber Utilizing Manganese in Metal-Insulator-Metal Structure,” *Scientific Reports*, vol. 8, p. 9162, 2018.
2. **Majid Aalizadeh**, A. E. Serebryannikov, A. Khavasi, G. A. E. Vandenbosch, and E. Ozbay, “Toward Electrically Tunable, Lithography-Free, Ultra-Thin Color Filters Covering the Whole Visible Spectrum,” *Scientific Reports*, vol. 8, p. 11316, 2018.
4. **Majid Aalizadeh**, A. Khavasi, B. Butun, and E. Ozbay, “Lithography-Free, Manganese-Based Ultra-Broadband Absorption Through Annealing-Based Deformation of Thin Layers into metal-air composites,” *Optics Letters*, Doc. ID 369951, 2019.
3. **Majid Aalizadeh**, A. Khavasi, A. E. Serebryannikov, G. A. E. Vandenbosch, and E. Ozbay, “A Route to Unusually Broadband Plasmonic Absorption Spanning from Visible to Mid-infrared,” *Plasmonics*, 2019.
5. **Majid Aalizadeh**, M. R. Tavakol, A. Khavasi, M. Yilmaz, and E. Ozbay, “Electromagnetic Field Tapering in the High-Roughness Substrates Coated by a Thin Film of Manganese: A Lithography-Free Ap-

proach to Ultra-Broadband, Wide-Angle, UV to FIR Perfect Absorption,” *Submitted*, 2019.

6. **Majid Aalizadeh**, A. E. Serebryannikov, G. A. E. Vandenbosch, and E. Ozbay, “High-efficiency, Ultra-wideband and Wide-angle Beam Deflection and Multi-functional Spatial Filtering with Dielectric Nanorods”, *To be submitted*, 2019.
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