Research article

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Bismuth-based metamaterials: from narrowband reflective color filter to extremely broadband near perfect absorber

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Abstract: In recent years, sub-wavelength metamaterials-based light perfect absorbers have been the subject of many studies. The most frequently utilized absorber configuration is based on nanostructured plasmonic metals. However, two main drawbacks were raised for this design architecture. One is the fabrication complexity and large scale incompatibility of these nano units. The other one is the inherent limitation of these common metals which mostly operate in the visible frequency range. Recently, strong interference effects in lithography-free planar multilayer designs have been proposed as a solution for tackling these drawbacks. In this paper, we reveal the extraordinary potential of bismuth (Bi) metal in achieving light perfect absorption in a planar design through a broad wavelength regime. For this aim, we adopted a modeling approach based on the transfer matrix method (TMM) to find the ideal conditions for light perfect absorption. According to the findings of our modeling and numerical simulations, it was demonstrated that the use of Bi in the metal-insulator-metal-insulator (MIMI) configuration can simultaneously provide two distinct functionalities; a narrow near unity reflection response and an ultra-broadband near perfect absorption. The reflection behavior can

be employed to realize additive color filters in the visible range, while the ultra-broadband absorption response of the design can fully harvest solar irradiation in the visible and near infrared (NIR) ranges. The findings of this paper demonstrate the extraordinary potential of Bi metal for the design of deep sub-wavelength optical devices.

Keywords: perfect absorber; metamaterials; color filter; solar irradiation; plasmonic resonance; impedance matching.

1 Introduction

Optical metamaterials have been the subject of intensive investigations for the design of highly efficient sub-wavelength devices in recent years. Strong light-matter interaction in properly designed nanostructures can be utilized to fully harvest the incident solar irradiation in a broad or narrow wavelength range. These metamaterial-based perfect absorbers can be employed in various applications. While these narrowband absorbers are of great interest for sensing and spectroscopy applications [1–7], several other areas such as photovoltaic and thermal photovoltaic [8–14] steam generation [15, 16] and photodetection [17] require the spectral broadness.

The common architecture for the design of light perfect absorbers is based on metal-insulator-metal (MIM) cavity. In this nanocavity design, the top layer is made of plasmonic metal nano units, while the bottom layer is an optically thick metal mirror. The common types of metals for this MIM cavity design are noble metals such as gold (Au), silver (Ag), and aluminum (Al). However, in general, the response of this type of cavity is narrow [2, 4, 6, 18–24]. To broaden the absorption bandwidth (BW) of this design, different strategies have been proposed. Multi-dimensional/multi-shape nano units [10, 25, 26–32], elongated designs [33], and replacing noble metals with lossy metals such as titanium (Ti) are examples of these

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approaches [34, 35]. However, most of these designs can only cover a part of the visible or near infrared (NIR) spectrum and their absorption BW is inherently restricted. In addition to this, electron beam lithography, which is not a large scale compatible fabrication technique, is involved in the fabrication route for all of these designs. Thus, this limits their further up-scaling.

To tackle this drawback, the concept of lithography-free multilayer planar perfect absorbers made of metal-insulator (MI) pairs has emerged [36–65]. The use of different metal types in MIM and metal-insulator-metal-insulator (MIMI) configurations has already been explored to achieve an ultra-broadband absorption BW. Increasing the number of pairs has been found as a proper way to further extend the operational BW of the design [52]. However, this leads to a new requirement where multiple deposition steps can affect the complexity and repeatability of the process. Moreover, even in this multilayer architecture, the BW is limited. For instance, it has been proven that the periodic arrangement of 16 pairs of Ni/SiO₂ and Ti/SiO₂ (where the metal layer thickness is in the order of 1–2 nm) could provide near unity light absorption up to 2.5 μm [52]. Therefore, together with configuration, a proper choice of material is also a prominent factor in obtaining a broad absorption band [66].

In this paper, we reveal the extraordinary potential of bismuth (Bi) metal in the design of light perfect absorbers. For this purpose, as an initial step, a modeling methodology based on the transfer matrix method (TMM) was developed for MIM and MIMI configurations to find the permittivity data of an ideal perfect absorber. According to the findings of this model, an ideal absorber should have a positive real part of the permittivity in longer wavelength values (i.e. the NIR spectrum). However, almost all metals have large negative real epsilon values in this frequency range. Hence, this limits their upper absorption edge and mitigates their further broadening. However, Bi has extraordinary permittivity data which is closely matched with the ideal absorber profile. Thus, the abovementioned limit is not applied for this metal. Inspired by these findings, we numerically investigated the absorption spectra of Bi-based MIM and MIMI cavities. In addition to their ultra-broadband absorption BW, Bi-based absorbers exhibit a narrowband high amplitude reflective response in the shorter wavelength values. This introduces a new functionality to our cavity design, which is highly efficient reflective color filtering. According to our results, the MIMI configuration has both a broader absorption BW and a narrower reflection width compared to that of the MIM cavity. To verify our simulation and theoretical expectations, different MIMI cavity designs

with different insulator layer geometries were fabricated. Optical characterizations of the fabricated designs are in line with our expectations where the Bi-based cavity can simultaneously operate as a narrowband perfect reflector and an ultra-broadband perfect absorber. To the best of our knowledge, this is the broadest reported absorption BW for a planar lithography-free multilayer design. As demonstrated in this study, the right choice of material could lead to a highly efficient optical device in dimensions much smaller than the wavelength.

2 Results and discussion

To begin with, as shown in Figure 1A, the ideal permittivity data was extracted for ideal MIM and MIMI light perfect absorbers. For this aim, as stated above, the TMM was utilized. The details of this modeling approach have already been provided in our previous studies [66]. In this modeling, the thickness of the bottom Ag metal layer is chosen as D_R , which is an optically thick mirror with no transmission and near unity reflection. The middle ideal absorber material has a thickness of D_M which is sandwiched with two symmetric insulator layers with a thickness of D_I . For this insulator layer, lithium fluoride (LiF) was chosen because it provides us the opportunity to deposit the whole structure in a single thermal evaporation step. Clearly, similar dielectrics such as Si, MgF₂, SiO₂ or TiO₂ can also be used as a dielectric layer, but they have generally high melting temperatures, and thermal evaporation is not a suitable technique for their deposition. Before analyzing the ideal conditions for obtaining near perfect absorption in these designs, it is better to revisit the permittivity data of common metals and compare it with that of Bi. Figure 1B–C depict the real and imaginary parts of the permittivity for different metals including Au, W, Ni, Ti, and Bi. For Bi, we experimentally extracted this permittivity profile, while for other metals, Palik's model was employed [67]. It can be clearly seen that, as we go toward longer wavelength values, the real permittivity data has an exponential drop towards negative values for all different metals. However, Bi showed an extraordinary permittivity response. It stayed in small negative values in visible and short NIR ranges and became positive in longer wavelengths. Moreover, it had smaller imaginary parts compared to other types of metals.

The ideal permittivity data for an absorption above 0.9 was extracted for different design geometries in MIM and MIMI configurations. These ideal regions are highlighted with brown (for the real part) and green (for the

Figure 1: The matching condition between Bismuth and ideal absorber.

(A) The schematic representation of metal-insulator-metal (MIM) and metal-insulator-metal-insulator (MIMI) cavity designs. The (B) real and (C) imaginary parts of permittivity for different types of metals, including Bi. The extracted ideal absorption regions for the real and imaginary parts of permittivity in an MIM cavity design for three different configurations of (D) $D_M = 5 \text{ nm}$ and $D_I = 120 \text{ nm}$, (E) $D_M = 20 \text{ nm}$ and $D_I = 120 \text{ nm}$, and (F) $D_M = 20 \text{ nm}$ and $D_I = 180 \text{ nm}$. The extracted ideal absorption regions for the real and imaginary parts of permittivity in an MIMI cavity design for three different configurations of (G) $D_M = 5 \text{ nm}$ and $D_I = 120 \text{ nm}$, (H) $D_M = 20 \text{ nm}$ and $D_I = 120 \text{ nm}$, and (I) $D_M = 20 \text{ nm}$ and $D_I = 180 \text{ nm}$.

imaginary part). For a specific wavelength, if the permittivity data of a metal is located within this area, it means that the structure can harvest above 90% of incident light. To understand the impact of different geometries in the overall absorption response of the system, we extracted the ideal data for different D_M and D_I values. These different combinations were extracted for the MIM case and the obtained ideal regions were compared with the permittivity data of Bi (see Figure 1D–F). As these panels clearly show, in ultrathin absorber thicknesses of $D_M = 5 \text{ nm}$, the matching could not be acquired for the imaginary part of epsilon. However, by increasing the thickness of the layer to 20 nm, the ideal region could be perfectly matched with that of Bi permittivity data for both the real and imaginary

parts, up to $2.5 \mu\text{m}$. At the same time, increasing the insulator layer thickness was used as a parameter to tune the optical response of the system. For $D_M = 20 \text{ nm}$, two different D_I values of 120 nm and 180 nm were studied in panels E and F. The ideal region for the imaginary part was almost the same for both cases, but the real part experienced a red shift, as expected. Thus, it is envisioned that the absorption upper edge cannot be altered by increasing the spacer layer. On the other end of the scale, we can see that for all cases, there was a mismatch in the visible part of the spectrum. The spectral position of this mismatch moved toward longer wavelengths as we increased the insulator layer thickness. In fact, this mismatch is located in the wavelength range where we have poor absorption

Figure 2: The parametric study on the cavity dimension to optimize light perfect absorption.

The absorption spectra of the metal-insulator-metal (MIM) design for different (A) D_M and (B) D_I values. The absorption spectra of the MIMI cavity for different (C) D_M and (D) D_I values. (E) The reflection spectra of the MIM and MIMI designs in the visible frequency range for three different D_I values of 110 nm, 170 nm, and 230 nm. (F) The absorption contour plot across the cavity for MIM and MIMI architectures for the case of $D_I = 110$ nm.

behavior and strong reflection response. The same parametric study was conducted for the MIMI case, as shown in Figure 1G–I. Similar behavior was recorded for the MIMI case, with the difference that the ideal region was wider compared with that of the MIM case. Moreover, the spectral width of the notch characteristic was narrower compared to that of the MIM design. Therefore, all these results tell us that the MIMI case has a better functionality compared to the MIM cavity. To check the validity of our modeling estimations, the numerical simulations, using a commercial finite-difference time-domain (FDTD) software package (Lumerical FDTD Solutions) [68], were further employed to investigate the absorption capability of the MIM and MIMI cavity designs. In the first step, the optimum D_M dimension was found for a fixed D_I value of 140 nm. As clearly seen from Figure 2A, the absorption response of the MIM design was quite weak for an ultrathin top layer thickness of 5 nm. This is in accordance with our theoretical findings. The increase in the top layer thickness made the absorption amplitude stronger and the BW wider. However, a broad dip in the absorption profile started to appear in the middle wavelength range (1–2 μm). To analyze these results, we reviewed the theoretical findings for the ideal model in Figure 2. Increasing the top layer thickness made the bottom limit of the tolerable region smaller for the imaginary part of epsilon. The same

behavior was recorded for the real part (compare the ideal regions in Figure 1D and E). Thus, while overall matching for both components was extended toward longer wavelength values, a mismatch started to appear in the real part for middle wavelength values. This mismatch shows itself as a dip in absorption. Therefore, a moderate thickness value should be chosen to have strong and nearly flat light absorption in a broad wavelength range. For this aim, the optimal value for D_M was picked as 20 nm, where the absorption profile stayed above 0.8 throughout the whole operation regime. Fixing the top layer thickness at this optimal amount, this time we investigated the impact of D_I thickness on the harvesting capability of the design. The sweep on D_I thickness was conducted from 110 nm to 230 nm with a step size of 30 nm. As we see in Figure 2B, moving toward thicker spacer layers, the response experienced a red shift, but the absorption stayed above 0.8 in the operation BW of the design. This was also in agreement with our theoretical estimations. A similar parametric study was applied for the MIMI configuration, as shown in Figure 2C and D. For this case, the optimal value for D_M was found to be 25 nm. However, the response in this case was stronger and the near unity absorption was satisfied in a broader wavelength range.

For instance, in the MIM and MIMI cases for $D_M = 20$ nm and $D_I = 140$ nm geometries, the absorption stayed above

0.8 throughout 0.63–2.45 μm and 0.56–2.59 μm , respectively. Moreover, the absorption profile for the MIMI case was quite flat and near unity.

Another feature that should be discussed in these figures is the visible response of these configurations. As can be clearly seen, a strong and narrow notch appeared for both configurations and the spectral position of this notch monolithically shifted toward longer wavelength values as we increased the spacer thickness. Considering the fact that this structure has no transmission, this notch behavior in the absorption profile is equivalent to a peak in the reflection response of the design. Therefore, this means that the structure is acting as a reflective red-green-blue (RGB) color filter. In a common planar lithography-free Fabry-Perot (FP) cavity design (such as MIM), the structure performs as a band-stop filter at the reflection mode that absorbs a narrow frequency range and reflects the rest of the spectrum [36, 37]. In these FP resonators, the RGB additive colors and their complementary subtractive ones of cyan-magenta-yellow can be obtained in transmission and reflection modes, respectively. Therefore, the generation of RGB additive colors is not possible in these designs and they can create these colors in the transmission spectra. However, due to the inherent loss of the materials, transmissive color filters are, in general, less efficient compared to reflective ones (generally limited to efficiencies below 0.7) [69]. However, the extraordinary optical response of Bi metal provides the opportunity to create these RGB colors in the reflection with reflection amplitudes as high as near unity. Thus, this structure will simultaneously act as an ultra-broadband perfect absorber and narrowband perfect reflector. To compare the reflective behavior of the MIM and MIMI designs, we drew the reflection spectra for three different D_i values with the optimal D_m for each case. As we can see from Figure 2E, the reflection peaks for both configurations had almost the same amplitude, but the spectral width for the MIMI cavity was narrower. This is actually beneficiary for color filtering application of these structures, where narrower peaks lead to more pure colors and less crosstalk. Hence, the MIMI cavity is not only better from an absorption broadness perspective, but it also provides narrower reflection peaks. The profile of the absorbed power versus the incident light wavelength is plotted across the cavity in Figure 2F. As we can see from this panel, the middle Bi layer became transparent into the incident wave in the resonance wavelength of the reflection peak. This transparency was retained in a narrower range for the MIMI cavity design, compared to that of the MIM case.

To verify our modeling and simulation results, the optimized MIMI configurations were fabricated using a

single thermal evaporation step. The bottom metal layer was Ag, the insulator layers were LiF, and the middle layer was Bi. Figure 3A shows the cross-sectional scanning electron microscopy image of the fabricated design. This image shows the successful formation of the MIMI cavity structure with an optimal Bi layer thickness of 24 nm, which is quite close to our intended goal. The bottom layer thickness was ~ 70 nm to act as a mirror in our operation wavelength. The LiF layer thicknesses were picked as 105 nm, 130 nm, 150 nm, 190 nm, and 220 nm to generate five different visible light colors; blue, cyan, green, orange, and red, respectively. The optical images of these five different samples are shown in Figure 3B. Use of the proper materials as insulators and metal layers can provide us the opportunity to fabricate a planar and spatially tunable color filter using a simple evaporation step. Getting a benefit from this property, we made a thickness variant multi-color high efficiency color filter design. The distance between the evaporation source and the sample corners was adjusted in a way that the insulator layer thickness linearly increased from one end to the other. The details of this fabrication are in the experimental section. The normal incident reflection spectra of the MIMI designs throughout the 0.35–4 μm range are depicted in Figure 3D–H. The reflection spectra of the structure for $D_i = 105$ nm show a reflection peak at 399 nm, corresponding to a blue color. The same structure shows reflection below 0.1 (corresponding to absorption above 0.9) across an ultra-broadband range spanning from 0.51 μm to 2.51 μm . For the case of 130 nm thick spacer layers, the reflection peak moved to 467 nm and the perfect absorption region (absorption above 0.9) spanned a 0.62–2.42 μm range. The absorption BW decreased for the other three colored samples, while the reflection peak monolithically red shifted. These experimental results are in great agreement with our modeling and simulation findings. For all colors, the reflection peak amplitude stayed above 0.8, which shows a high efficiency color generation out of a planar design. Thus, as stated above, the cavity architecture represents both ultra-broadband perfect absorption and spectrally selective narrowband perfect reflection.

To understand the angular response of the cavity, the absorption spectra of the design were measured under p and s polarized light for three different incident angles of $\theta = 30^\circ$, $\theta = 45^\circ$, and $\theta = 60^\circ$. The angular reflection response of the cavity is depicted in Figure 4A–E. These results were measured in the range of 0.35–2 μm using V-Vase ellipsometry equipment. The overall trend for all five different samples was similar. For both polarizations, the increase in incident light angle led to a blue shift in the reflection peak of the structure. This was expected, considering the fact

Figure 3: The measurement results for different colored MIMI cavities.

(A) The cross-sectional scanning electron microscopy (SEM) image of the fabricated Ag-LiF-Bi-LiF MIMI cavity. (B) The optical images of fabricated samples with different insulator layer thicknesses of 105 nm, 130 nm, 150 nm, 190 nm, and 220 nm. Each of these thicknesses leads to generation of a specific color in the reflection mode. (C) A monolithically designed spatially variant color filter by linear variation of insulator thickness across the sample, by adjusting the geometrical parameters of the deposition system. The normal incident absorption spectra of the MIMI cavity for different D_1 values of (D) 105 nm (blue), (E) 130 nm (cyan), (F) 150 nm (green), (G) 190 nm (orange), and (H) 220 nm (red). In all samples, the Bi layer thickness was 25 nm, which is the optimal value.

that the optical path length of the light increases as light impinges the surface in a wider view angle. This could be mitigated by the use of a higher refractive index medium as a spacer [69]. Based on Snell's law, a higher refractive index makes the incident light closer to the normal direction and this consequently reduces the angular dependency of the result. Moreover, the absorption strength of the design is retained high for all these wide incident angles. For p polarized excitation, the absorption amplitude became larger toward larger angles, while for s polarized light, the perfect absorption capability gradually diminished. This is due to different reflection coefficients seen by incident light in

these two different incident light polarizations. It should be mentioned that Bi has a low melting point, which limits its thermal stability and its potential use in high temperature applications, such as thermal photovoltaics. However, from an optical perspective, it has many advantages compared to other noble metals. As explained in the theoretical section, the absorption BW in other noble metal-based absorbers is much narrower and solar irradiation cannot be fully harvested. However, the broadband absorption response of the Bi layer could be of great interest for energy conversion applications such as hot electron-based photodetection, photovoltaics and photoelectrochemical water splitting.

Figure 4: Angular absorption response of the MIMI cavities.

The angular response of (A) blue, (B) cyan, (C) green, (D) orange, and (E) red colored filters under different incidence angles (i.e. 30°, 45°, and 60°) and polarizations (s and p). In all samples, the reflection gets a blue shift as we go toward wider viewing angles.

The temperature levels in these applications are generally much below the melting point of Bi metal. Moreover, the color reflective behavior can be employed in filtering and colorimetric sensing, which are again low thermal budget applications. Moreover, Bi is a low-cost earth-abundant element, unlike other precious and rare metals such as gold. Our group is currently working on the development of Bi-based optoelectronic devices for the abovementioned applications.

3 Conclusions

In this study, we revealed the extraordinary optical response of Bi metal in the design of deep sub-wavelength resonant cavities. Using a modeling approach based on the TMM, we first found the ideal condition for light perfect absorption. While all other common metals can only satisfy this ideal condition in the visible and short NIR range, Bi is perfectly matched to it in an ultra-broadband frequency range. Additionally, the structure was predicted to act as a narrowband perfect reflector in the visible range. Thus, a similar cavity can propose two distinct functionalities. Afterward, numerical simulations were employed to find the optimal geometries for achieving the best performance. The characterization results of the fabricated samples show that the MIMI cavities have

both ultra-broadband near perfect absorption and narrowband perfect reflection, where the spectral position of the reflection peak can be easily tuned by changing the spacer insulator layer thickness. It was found that this planar structure can absorb more than 90% of incident light in an extremely broadband regime spanning from 0.51 μm to 2.51 μm . To the best of our knowledge, this is the highest absorption BW reported compared to any MI pair-based planar design. Moreover, the structure can create RGB colors in the reflection mode with high efficiency, high purity, and low crosstalk, which generally cannot be achieved using common FP-based designs. All in all, the findings of this study demonstrate a high potential for Bi metal to be used in optical devices. This paper shows the importance of the material selection for the design of deep sub-wavelength optical cavity structures.

4 Experimental

4.1 Device fabrication

To fabricate this structure, first, the Si (Sigma-Aldrich; Munich, Germany) wafer was diced into small pieces. Then, the samples were placed inside a Piranha solution for a duration of 10 min. Finally, hydrofluoric acid (HF) (Sigma-Aldrich; Munich, Germany) was utilized to remove the

native oxide layer. A thermal evaporator machine was utilized to coat the Ag, Bi, and LiF (Kurt J. Lesker; Toronto, ON, Canada) layers at the desired thicknesses, using the thermal evaporation technique (VAKSIS; Ankara, Turkey). The deposition rate was set at around 2 Å/s for all layers and the chamber pressure was kept below 5×10^{-6} Torr throughout the deposition process. To obtain a linearly variant color filter design, the sample was brought closer to the evaporation source. The position of the sample was adopted in a way that one side of it was at an 8 cm distance from the evaporation source, while the other side was ~ 12 cm away.

4.2 Optical characterization

To measure the normal incidence of light reflection from the structure, in a wavelength range of $0.35\text{--}1.1 \mu\text{m}$, we used an in-house setup comprising a halogen lamp as the incident light source, where this source was integrated into a microscope to collect the reflected light from the surface. This collected light intensity in the microscope was entered into a spectrometer (Newport Corporation; Irvine, CA, USA) (Newport OSM2). In all of the measurements, the obtained reflection values were normalized with the reflection data from a thick Al-coated sample (which had near 100% reflection in our desired frequency range). Finally, a personal computer was utilized to extract and monitor the data. For the longer wavelength values (the NIR range), we utilized a Fourier-transform infrared spectroscopy (FTIR) (Hyperion 2000; Bruker, Billerica, MA, USA) system. The numerical aperture of the FTIR equipment was ~ 0.6 with a half-cone angle of 37° and a spot size of $250 \mu\text{m}$. The homemade reflection setup had a numerical aperture of 0.22, with a half-cone angle of 13° . For the FTIR measurement, the spectra were recorded in the reflection mode of a Hyperion microscope using a liquid nitrogen-cooled MCT-D316 (mercury cadmium telluride) detector and a global light source. The obtained data is renormalized by the reflection response of an Al mirror. In addition, the reflection of the MIM samples at different light incident angles was measured utilizing a spectroscopic ellipsometer tool (V-Vase; J.A. Wollam Co. Inc.; Lincoln, NE, USA) at two different s and p polarizations. The light angles of incidence were chosen to be 30° , 45° , and 60° . Moreover, the permittivity data of Bi was obtained using this equipment. To extract the permittivity function of the Bi thin film, we first measured the angular response of the layer using ellipsometry. Then, the acquired data was fitted with a linear sum of the Drude model and the Kramers-Kronig Lorentz oscillator. We fitted the selected dielectric functions by using a linear sum of a Drude dielectric function and Kramers-Kronig Lorentz oscillators accounting for

interband transitions. Two unknowns in the Drude model, electron density and collision frequency, were found from the literature [70]. Later, Lorentz oscillators were added one by one and the parameters were tuned until we got a reasonable fit. Finally, all oscillator-related parameters were fixed in their optimal values and this time, the Drude-related coefficients were tuned to maximize the fitting match. It should be mentioned that the overall number of oscillators for the abovementioned fit was six. Moreover, to extract the LiF permittivity values, we used ellipsometry measurements as we did for Bi. The obtained data was fitted into the Cauchy model.

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References

- [1] Liu N, Mesch M, Weiss T, Hentschel M, Giessen H. Infrared perfect absorber and its application as plasmonic sensor. *Nano Lett* 2010;10:2342–8.
- [2] Yong Z, Zhang S, Gong C, He S. Narrow band perfect absorber for maximum localized magnetic and electric field enhancement and sensing applications. *Sci Rep* 2016;6:24063.
- [3] Wu D, Li R, Liu Y, et al. Ultra-narrow band perfect absorber and its application as plasmonic sensor in the visible region. *Nanoscale Res Lett* 2017;12:427.
- [4] Wu D, Liu Y, Li R, et al. Infrared perfect ultra-narrow band absorber as plasmonic sensor. *Nanoscale Res Lett* 2016;11:483.
- [5] Lu X, Zhang L, Zhang T. Nanoslit-microcavity-based narrow band absorber for sensing applications. *Opt Express* 2015;23:20715–20.
- [6] Chang W, Won J, Slaughter LS, Link S. Plasmonic nanorod absorbers as orientation sensors. *Proc Natl Acad Sci USA* 2010;107:2781–6.
- [7] Lu X, Wan R, Zhang T. Metal-dielectric-metal based narrow band absorber for sensing applications. *Opt Express* 2015;23:29842–7.
- [8] Rinnerbauer V. Metallic photonic crystal absorber-emitter for efficient spectral control in high-temperature solar thermophotovoltaics. *Adv Energy Mater* 2014;4:1–10.
- [9] Wang H, Chen Q, Wen L, Song S, Hu X, Xu G. Titanium-nitride-based integrated plasmonic absorber/emitter for solar thermophotovoltaic application. *Photonic Res* 2015;3:329–34.
- [10] Wang H, Wang L. Perfect selective metamaterial solar absorbers. *Opt Express* 2013;21:13311–9.
- [11] Rephaeli E, Fan S. Absorber and emitter for solar thermophotovoltaic systems to achieve efficiency exceeding the Shockley-Queisser limit. *Opt Express* 2009;17:15145–59.

- [12] Wu C, Neuner B, John J, et al. Metamaterial-based integrated plasmonic absorber/emitter for solar thermo-photovoltaic systems. *J Opt* 2012;14:024005.
- [13] Farhat M, Cheng T-C, Le KQ, Cheng MMC, Bağcı H, Chen P-Y. Mirror-backed dark alumina: a nearly perfect absorber for thermoelectronics and thermophotovoltaics. *Sci Rep* 2016;6:19984.
- [14] Ghobadi TGU, Ghobadi A, Ozbay E, Karadas F. Strategies for plasmonic hot-electron-driven photoelectrochemical water splitting. *ChemPhotoChem* 2018;2:1–23.
- [15] Zhou L, Tan Y, Ji D, et al. Self-assembly of highly efficient, broadband plasmonic absorbers for solar steam generation. *Sci Adv* 2016;2:e1501227.
- [16] Bae K, Kang G, Cho SK, Park W, Padilla WJ, Kim K. Flexible thin-film black gold membranes with ultrabroadband plasmonic nanofocusing for efficient solar vapour generation. *Nat Commun* 2015;6:1–9.
- [17] Li W, Valentine J. Metamaterial perfect absorber based hot electron photodetection. *Nano Lett* 2014;14:3510–4.
- [18] Moreau A, Ciraci C, Mock JJ, et al. Controlled-reflectance surfaces with film-coupled colloidal nanoantennas. *Nature* 2012;492:86–90.
- [19] Cattoni A, Ghenuche P, Decanini D, Chen J, Pelouard J. $\lambda/1000$ Plasmonic nanocavities for biosensing fabricated by soft UV nanoimprint lithography. *Nano Lett* 2011;11:3557–63.
- [20] Zhang N, Dong Z, Ji D, et al. Reversibly tunable coupled and decoupled super absorbing structures. *Appl Phys Lett* 2016;108:091105.
- [21] Landy NI, Sajuyigbe S, Mock JJ, Smith DR, Padilla WJ. Perfect metamaterial absorber. *Phys Rev Lett* 2008;100:207402.
- [22] Mandal P. Plasmonic metamaterial perfect absorber at visible frequency: design rule. *J Nanophotonics* 2017;11:036003.
- [23] Wang B, Huang W-Q, Wang L-L. Ultra-narrow terahertz perfect light absorber based on surface lattice resonance of a sandwich resonator for sensing applications. *RSC Adv* 2017;7:42956–63.
- [24] Kim J, Han K, Hahn JW. Selective dual-band metamaterial perfect absorber for infrared stealth technology. *Sci Rep* 2017;7:6740.
- [25] Liu X, Starr T, Starr AF, Padilla WJ. Infrared spatial and frequency selective metamaterial with near-unity absorbance. *Phys Rev Lett* 2010;104:207403.
- [26] Nielsen MG, Pors A, Albrechtsen O, Bozhevolnyi SI. Efficient absorption of visible radiation by gap plasmon resonators. *Opt Express* 2012;20:13311–9.
- [27] Ming X, Tan Q. Design method of a broadband wide-angle plasmonic absorber in the visible range. *Plasmonics* 2017;12:14–17.
- [28] Aydin K, Ferry VE, Briggs RM, Atwater HA. Broadband polarization-independent resonant light absorption using ultrathin plasmonic super absorbers. *Nat Commun* 2011;2:517.
- [29] Kenney M, Grant J, Shah YD, Escorcia-carranza I, Humphreys M, Cumming DRS. Octave-spanning broadband absorption of terahertz light using metasurface fractal-cross absorbers. *ACS Photonics* 2017;4:2604–12.
- [30] Duan X, Chen S, Liu W, Cheng H, Li Z. Polarization-insensitive and wide-angle broadband nearly perfect absorber by tunable planar metamaterials in the visible regime. *J Opt* 2014;16:125107.
- [31] Azad AK, Kort-Kamp WJM, Sykora M, et al. Metasurface broadband solar absorber. *Sci Rep* 2016;6:20347.
- [32] Mudachathi R, Tanaka T. Broadband plasmonic perfect light absorber in the visible spectrum for solar cell applications. *Adv Nat Sci Nanosci Nanotechnol* 2018;9:015010.
- [33] Ghobadi A, Hajian H, Gokbayrak M, et al. Visible light nearly perfect absorber : an optimum unit cell arrangement for near absolute polarization insensitivity. *Opt Express* 2017;25:20256–65.
- [34] Ding F, Dai J, Chen Y, Zhu J, Jin Y, Bozhevolnyi SI. Broadband near-infrared metamaterial absorbers utilizing highly lossy metals. *Sci Rep* 2016;6:39445.
- [35] Ghobadi A, Hajian H, Rashed AR, Butun B, Ozbay E. Tuning the metal filling fraction in metal-insulator-metal ultra-broadband perfect absorbers to maximize the absorption bandwidth. *Photonics Res* 2018;6:168–76.
- [36] Park CS, Shrestha VR, Lee SS, Choi DY. Trans-reflective color filters based on a phase compensated etalon enabling adjustable color saturation. *Sci Rep* 2016;6:25496.
- [37] Li Z, Butun S, Aydin K. Large-area, lithography-free super absorbers and color filters at visible frequencies using ultrathin metallic films. *ACS Photonics* 2015;2:183–8.
- [38] Zhao Y, Zhao Y, Hu S, et al. Artificial structural color pixels: a review. *Materials (Basel)* 2017;10:944.
- [39] Yoon Y-T, Lee S-S. Transmission type color filter incorporating a silver film based etalon. *Opt Express* 2010;18:5344–9.
- [40] Zhao D, Meng L, Gong H, et al. Ultra-narrow-band light dissipation by a stack of lamellar silver and alumina. *Appl Phys Lett* 2014;104:221107.
- [41] Lee JY, Lee KT, Seo S, Guo LJ. Decorative power generating panels creating angle insensitive transmissive colors. *Sci Rep* 2014;4:4192.
- [42] Mirshafieyan SS, Gregory DA. Electrically tunable perfect light absorbers as color filters and modulators. *Sci Rep* 2018;8:1–9.
- [43] Peng H, Luo Y, Ying X, et al. Broadband and highly absorbing multilayer structure in mid-infrared. *Appl Opt* 2016;55:8833–8.
- [44] Li Z, Palacios E, Butun S, Kocer H, Aydin K. Omnidirectional, broadband light absorption using large-area, ultrathin lossy metallic film coatings. *Sci Rep* 2015;5:15137.
- [45] Ghobadi A, Dereshgi SA, Butun B, Ozbay E. Ultra-broadband asymmetric light transmission and absorption through the use of metal free multilayer capped dielectric microsphere resonator. *Sci Rep* 2017;7:14538.
- [46] Mattiucci N, Bloemer MJ, Akozbek N, D'Aguzzo G. Impedance matched thin metamaterials make metals absorbing. *Sci Rep* 2013;3:1–11.
- [47] Chirumamilla M, Roberts AS, Ding F, et al. Multilayer tungsten-alumina-based broadband light absorbers for high-temperature applications. *Opt Mater Express* 2016;6:2704–14.
- [48] Mao K, Shen W, Yang C, et al. Angle insensitive color filters in transmission covering the visible region. *Sci Rep* 2016;6:19289.
- [49] Deng H, Li Z, Stan L, Rosenmann D, Czaplewski D. Broadband perfect absorber based on one ultrathin layer of refractory metal. *Opt Lett* 2015;40:2592–5.
- [50] Kajtár G, Kafesaki M, Economou EN, Soukoulis CM. Theoretical model of homogeneous metal–insulator–metal perfect multiband absorbers for the visible spectrum. *J Phys D Appl Phys* 2016;49:055104.
- [51] Ding F, Mo L, Zhu J, Sailing He S. Lithography-free, broadband, omnidirectional, and polarization-insensitive thin optical absorber. *Appl Phys Lett* 2015;106:061108.

- [52] Zhong YK, Lai YC, Tu MH, et al. Omnidirectional, polarization-independent, ultra-broadband metamaterial perfect absorber using field-penetration and reflected-wave-cancellation. *Opt Express* 2016;24:A832–45.
- [53] Zhong YK, Fu SM, Ju NP, Tu M-H, Chen B-R, Lin A. Fully planarized perfect metamaterial absorbers with no photonic nanostructures. *IEEE Photonics J* 2016;8:2200109.
- [54] Fu SM, Zhong YK, Tu MH, Chen BR, Lin A. A fully functionalized metamaterial perfect absorber with simple design and implementation. *Sci Rep* 2016;6:36244.
- [55] Ghobadi A, Dereshgi SA, Hajian H, Bozok B, Butun B, Ozbay E. Ultra-broadband, wide angle absorber utilizing metal insulator multilayers stack with a multi-thickness metal surface texture. *Sci Rep* 2017;7:4755.
- [56] Ghobadi A, Hajian H, Dereshgi SA, Bozok B, Butun B, Ozbay E. Disordered nanohole patterns in metal-insulator multilayer for ultra-broadband light absorption: atomic layer deposition for lithography free highly repeatable large scale multilayer growth. *Sci Rep* 2017;7:15079.
- [57] Dereshgi SA, Ghobadi A, Hajian H, Butun B, Ozbay E. Ultra-broadband, lithography-free, and large-scale compatible perfect absorbers: the optimum choice of metal layers in metal-insulator multilayer stacks. *Sci Rep* 2017;7:14872.
- [58] Ghobadi A, Hajian H, Soydan MC, Butun B, Ozbay E. Lithography-free planar band-pass reflective color filter using series connection of cavities. *Sci Rep* 2019;9:1–11.
- [59] Park CS, Shrestha VR, Lee SS, Kim ES, Choi DY. Omnidirectional color filters capitalizing on a nano-resonator of Ag-TiO₂-Ag integrated with a phase compensating dielectric overlay. *Sci Rep* 2015;5:8467.
- [60] Lee K-T, Seo S, Lee JY, Guo LJ. Strong resonance effect in a lossy medium-based optical cavity for angle robust spectrum filters. *Adv Mater* 2014;26:6324–8.
- [61] Han JH, Kim D-Y, Kim D, Choi KC. Highly conductive and flexible color filter electrode using multilayer film structure. *Sci Rep* 2016;6:29341.
- [62] Lee KT, Seo S, Yong Lee J, Jay Guo L. Ultrathin metal-semiconductor-metal resonator for angle invariant visible band transmission filters. *Appl Phys Lett* 2014;104:231112.
- [63] Yang C, Shen W, Zhang Y, et al. Compact multilayer film structure for angle insensitive color filtering. *Sci Rep* 2015;5:9285.
- [64] Lee KT, Han SY, Park HJ. Omnidirectional flexible transmissive structural colors with high-color-purity and high-efficiency exploiting multicavity resonances. *Adv Opt Mater* 2017;5:1700284.
- [65] Li Z, Butun S, Aydin K. Lithography-free transmission filters at ultraviolet frequencies using ultra-thin aluminum films. *J Opt* 2016;18:065006.
- [66] Ghobadi A, Hajian H, Butun B, Ozbay E. Strong light-matter interaction in lithography-free planar metamaterial perfect absorbers. *ACS Photonics* 2018;5:4203–21.
- [67] Palik ED. Handbook of optical constants of solids. Vol. 3. San Diego, USA, Academic Press, 1998.
- [68] Lumerical Solutions, Inc. Available at: <http://www.lumerical.com/tcad-products/fdtd/>.
- [69] Ji C, Lee K, Xu T, Zhou J, Park HJ, Guo LJ. Engineering light at the nanoscale : structural color filters and broadband perfect absorbers. *Adv Opt Mater* 2017;5:1700368.
- [70] Claessen LM, Jansen AGM, Wyder P. Plasma resonances in thin Bi films. *Phys Rev B* 1986;33:7947–55.