ADVANCED OPTICAL MATERIALS

www.advopticalmat.de

Designer Perfect Light Absorption Using Ultrathin Lossless Dielectrics on Absorptive Substrates

*Mohamed ElKabbash, Shamreen Iram, Theodore Letsou, Michael Hinczewski, and Giuseppe Strangi**

Optical absorbers comprised of an ultrathin lossy dielectric film on an opaque metallic substrate are an attractive alternative to lithographically intense metamaterial and nanoplasmonic optical absorbers as they allow for large-scale, cost-effective fabrication. However, requiring that the dielectric is lossy and the metallic substrate is highly reflective but not a perfect electric conductor (PEC) limits the wavelength range and materials that can be used to realize strong to perfect light absorption. In this work, we theoretically and experimentally investigate light absorption using ultrathin lossless dielectric films. By choosing proper lossless ultrathin dielectrics and substrates, iridescence free, perfect light absorption is possible over the visible, near infrared (NIR), and short-wave infrared (SWIR) wavelength ranges with designer absorption properties. The proposed class of ultrathin film absorbers relaxes many constraints on the type of materials used to realize perfect light absorption. The flexibility of our design makes it relevant for many applications specifically in structural coloring, selective thermal emission, thermo-photovoltaics, photo-thermoelectric generation, and gas sensing.

Optical absorbers, materials with absorptance A that enable strong ($A > 0.9$) to perfect ($A > 0.99$) light absorption, have many applications in thermo-photovoltaics, photodetectors, stealth technologies, thermal emission, optical switches, hybrid solar converters, and structural coloring.[1–6] When light is critically coupled to an optical resonator, light is perfectly absorbed. Critical light coupling takes place when the resonator absorption and reflection rates are equal.[7–10] A monolithic medium, that is, comprised of a single material, can realize perfect light absorption if the medium supports strong optical resonances due to the excitation of surface plasmon polaritons in metallic nanostructures[11] or via impedance matching using metamaterials.[12] However, the intense nanolithography required to realize controlled monolithic perfect absorbers in the optical regime can dramatically limit their practical applications.

Dr. Md. ElKabbash, S. Iram, T. Letsou, Prof. M. Hinczewski, Prof. G. Strangi
Department of Physics
Case Western Reserve University
10600 Euclid Avenue, Cleveland, OH 44106, USA
E-mail: gxs284@case.edu

Prof. G. Strangi
CNR-NANOTEC Istituto di Nanotecnologia and Department of Physics
University of Calabria
87036 Rende, Italy

DOI: 10.1002/adom.201800672

Is it possible to realize strong light absorption (SLA) using a smooth, unpatterned substrate (**Figure 1**a)? Figure 1b shows the calculated absorptance of TE-polarized light (λ = 700 nm) normally incident from air on an infinitely thick substrate for different values of the real (n_S) and imaginary (k_S) parts of the complex refractive index $\tilde{n}_S = n_S + ik_S$.[13] For values of approximately n_S = [0.5, 2] and k_s = [0.01, 1], SLA is possible. The optical constants of several materials are shown in Figure 1a and the corresponding absorptance values closely agree with the absorptance measured at 700 nm for these materials.[14] As can be seen from the figure, none of these materials lie in the SLA region for 700 nm, and in general it is difficult to find a material in bulk form that exhibits SLA in the optical range with air as an incident medium (see the Supporting Information). The lack of SLA in bulk materials is due to the strong impedance mismatch between the incidence medium and the absorbing medium.[15]

Adding an ultrathin dielectric coating with high losses on a reflective opaque substrate enabled strong to perfect light absorption in the visible, NIR, and IR ranges.[7,8,16] Critical light coupling in thin films takes place due to amplitude splitting destructive interference,[17] such that light is entirely trapped inside the resonator and is dissipated due to the existence of losses. Accordingly, two conditions must be simultaneously satisfied to realize perfect absorption; the interfering waves must be out of phase (phase condition), and the out-of-phase waves must be of equal amplitude (amplitude condition) (see for more details ref. [7]). Satisfying the amplitude condition is straightforward in asymmetric Fabry–Perot metal–dielectric–metal cavities as the top and bottom metal layers are highly reflective.[5,10] However, it is more difficult to satisfy the amplitude condition using a dielectric coating on a metallic substrate given that dielectrics do not strongly reflect light. On the other hand, a lossy dielectric dampens the magnitude of the partially reflected waves from the highly reflective metallic substrate which satisfies the amplitude condition. However, requiring a lossy dielectric limits the materials and spectral range where SLA is possible as most dielectrics are lossless in the optical range. For instance, although Ge is used as a lossy dielectric for perfect light absorption in the visible,[16] it weakly absorbs in the NIR. Furthermore, in order to satisfy the phase condition

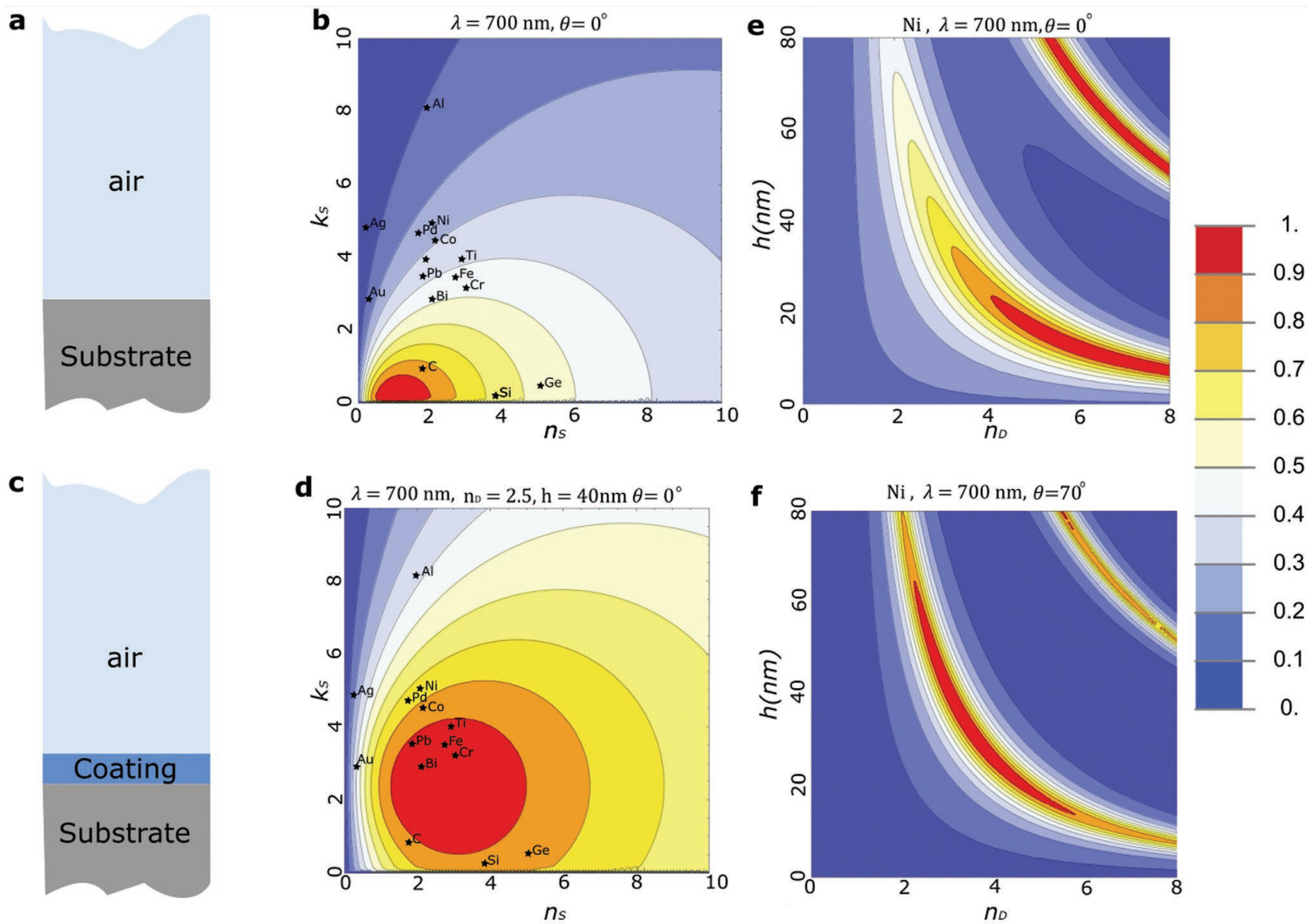

Figure 1. a) A schematic of homogeneous infinite substrate with a complex refractive index $\tilde{n}_S = n_S + ik_S$. b) The calculated absorptance of normally incident light with $\lambda = 700$ nm for the infinite substrate depicted in (a), the index $\tilde{n}_S$ of different materials is marked. Known materials do not exhibit strong light absorption (SLA). c) A schematic of a homogeneous ultrathin lossless dielectric coating on an infinite substrate. d) The calculated absorptance of normally incident light on the dielectric-substrate metasurface for different values of n_S and k_S. The range where strong to perfect light absorption takes place increased and includes several materials. e) For a Ni substrate, the absorptance of a given lossless dielectric is shown as a function of its thickness h and its refractive index n_D. For Ni, SLA is not possible for $n_D < 4$. f) The absorptance of a lossless dielectric on Ni substrate at higher incident angle (70°) is shown. SLA is possible for $n_D \approx 2$.

using an ultrathin dielectric coating with thickness h and refractive index n_D such that $h < \lambda/4n_D$ (as opposed to conventional antireflective coatings which requires $h = \lambda/4n_D$) the metallic substrate cannot behave as a perfect electric conductor (PEC) in order to provide a nontrivial phase shift (ϕ) upon reflection ($\phi \neq 0$ or π).[16] Highly reflective metallic substrates behave as PECs with a π reflection phase shift at frequencies lower than the visible which imposes further limitations on the design of ultrathin absorbers.

In this work, we study an alternative strategy to realize perfect light absorption. We show that iridescence free, strong to perfect light absorption is possible using lossless dielectrics with thickness $h < \lambda/(4n_D)$ on an absorptive substrate. We numerically and theoretically investigate the conditions required in the lossless dielectric and the absorptive substrate for SLA to take place. We show that the losses occur predominantly within few nanometers of the metallic substrate. The proposed design requires few conditions which enabled us to experimentally demonstrate light absorption across a large wavelength range using an ultrathin dielectric film on the same substrate.

Adding a lossless dielectric coating on an infinitely thick substrate can modify the absorption properties of the system (Figure 1c). If the dielectric is very thin, that is, $h \ll \lambda$, an equivalent semi-infinite medium can be defined that has the same absorptance as the combined thin-film/substrate structure. Accordingly, the thin-film/substrate structure can be regarded as a metasurface with controllable complex refractive indices.[18] The modified absorptance as a function of the substrate optical constants (n_S and k_S) is shown in Figure 1d for a lossless dielectric coating with $h = 40$ nm and $n_D = 2.5$. The absorptance did not increase significantly for substrates with high reflectance, for example, Al, Au, and Ag. On the other hand, the absorptance significantly increased for many substrates with higher absorptance, for example, Ni, Pd, Ti, Fe, Pb, Cr, Bi, and Co. Accordingly, SLA is possible for lossless dielectrics with $h < \lambda/4n_D$ (here $h \approx \lambda/7n_D$) on a variety of substrates. Note that the absorptance did not increase significantly for substrates with significant absorptance ($A > 0.5$), for example, C, Si, and Ge. Accordingly, having an absorptive substrate per se is insufficient to realize SLA without careful investigation of the dielectric coating parameters contrary to what is indicated in ref. [7]. The necessary balance between n_D and the absorption of the substrate is because the amplitude condition requires balancing the amplitude of partially reflected waves from the air–dielectric film interface and the film–substrate interface as we will demonstrate analytically shortly. Accordingly, SLA takes place for highly absorptive substrates even for dielectric superstrates with low refractive index (i.e., low reflectance) (see the Supporting Information).

For a given substrate, what are the conditions required in the lossless dielectric superstrate to realize SLA? Figure 1e shows the absorptance of a metamaterial comprised of an Ni substrate and a dielectric superstrate with different values for n_D and h at normal incidence. A low n superstrate, for example, $n_D = 1.5$ cannot achieve SLA regardless of its thickness which means that there is no SLA whether the phase condition is satisfied or not. On the other hand, for a dielectric with moderately high n_D, for example, $n_D = 2.5$, the absorptance is higher and reaches SLA at higher angles of incidence as shown in Figure 1f. Accordingly, by appropriately choosing the dielectric film/substrate metamaterial, we can control the wavelength and angular ranges where SLA takes place. The large variety of superstrate–substrate combinations and the flexibility to realize absorption with designer properties highlight the relevance of the introduced light absorber.

The conditions for perfect absorption in our system can be determined analytically using the standard transfer matrix technique to calculate the complex Fresnel reflection coefficient. Consider a superstrate with refractive index n_0, a lossless dielectric layer with thickness h and refractive index n_D, and a lossy substrate with refractive index $n_S + ik_S$. Perfect light absorption corresponds to the real and imaginary parts of the numerator of Fresnel reflection coefficient r simultaneously equal to zero. The general conditions for this occurring when light is incident on the system at angle θ do not have a simple analytical form. However, the conditions can be simplified and expressed in closed form in the limit where $|n_0 \sin\theta/(n_s + ik_s)|^2 \ll 1$. This limit turns out to be valid for all the cases of practical interest that we discuss, and in particular is exactly satisfied at normal incidence where $\theta = 0$. The resulting conditions for perfect absorption can be written as two equations for h and k_S that must be simultaneously satisfied.

For p-polarized incident light, the two equations have the form

$$h = \frac{\lambda}{2\pi\sqrt{n_D^2 - n_0^2\sin^2(\theta)}} \tan^{-1}\left(\sqrt{\frac{n_D^4\cos(\theta)\left(\cos(\theta)n_S - n_0\right)}{n_0\left(n_D^4\cos(\theta) - n_0 n_S\left(n_D^2 - n_0^2\sin^2(\theta)\right)\right)}}\right) \tag{1}$$

$$k_s = \sqrt{\frac{\left(n_S/n_0 - \sec(\theta)\right)\left(n_D^4\cos(\theta) - n_0 n_S\left(n_D^2 - n_0^2\sin^2(\theta)\right)\right)}{n_D^2 - n_0^2\sin^2(\theta)}} \tag{2}$$

and for s-polarized incident light

$$h = \frac{\lambda}{2\pi\sqrt{n_D^2 - n_0^2\sin^2(\theta)}} \tan^{-1}\left(\sqrt{\frac{\left(n_D^2\sec(\theta) - n_0^2\sin(\theta)\tan(\theta)\right)\left(n_S - n_0\cos(\theta)\right)}{n_0\left(n_D^2 - n_0\left(n_0\sin^2(\theta) + \cos(\theta)n_S\right)\right)}}\right) \tag{3}$$

$$k_S = \sqrt{\left(n_S/n_0 - \cos\theta\right)\left(n_D^2\sec(\theta) - n_0\left(n_0\sin(\theta)\tan(\theta) + n_S\right)\right)} \tag{4}$$

At normal incidence, $\theta = 0$, the above conditions become identical for s and p polarizations, simplifying to

$$h = \frac{\lambda}{2\pi n_D}\tan^{-1}\left(n_D\sqrt{\frac{n_S - n_0}{n_0\left(n_D^2 - n_0 n_S\right)}}\right) \tag{5}$$

$$k_S = \sqrt{\left(n_S/n_0 - 1\right)\left(n_D^2 - n_0 n_S\right)} \tag{6}$$

The last expression can also be inverted to give n_D in terms of the k_s and the other constants and is given by

$$n_D = \sqrt{n_0\left(\frac{k_S^2}{n_S - n_0} + n_S\right)} \tag{7}$$

For $k_S = 0$, in the absence of substrate absorption, we see that $n_D = \sqrt{n_0 n_S}$ and $h = \lambda/4\, n_D$, that is, we recover the conditions of dielectric antireflective coatings. As we increase k_S from zero, keeping all other optical constants fixed, n_D must increase to satisfy the perfect absorption condition, that is, $n_D > \sqrt{n_0 n_S}$ and the dielectric film thickness must decrease, that is, $h < \lambda/4n_D$, relative to the conventional antireflective condition. This conclusion supports our previous claim that for a highly reflective substrate, it is necessary to have a high index dielectric film to satisfy the amplitude condition. We provide a numerical example in Section S2 (Supporting Information). Note that since the arctan expression is always $\leq\pi/2$, the dielectric film thickness is constrained by $h \leq \lambda/\left(4\sqrt{n_D^2 - n_0^2\sin^2(\theta)}\right)$. For a given substrate and incidence angle, the above results allow one to choose a dielectric film with appropriate index n_D to satisfy k_s condition. Plugging the desired value of n_D into the corresponding h equation yields the required film thickness. In the absence of a dielectric film coating, achieving perfect absorption puts a much more stringent requirement on the substrate: the conditions become $k_S = 0$ and $n_S = n_0\sec(\theta)$ (for p polarization), $n_S = n_0\cos(\theta)$ (for s polarization) (for detailed derivation of Equations (1)–(4), see the Supporting Information).

To experimentally demonstrate the properties of the introduced optical absorber, we fabricated two sets of thin-film structures comprised of a lossless dielectric (TiO_2) on 90 nm Ag and 90 nm Ni substrates, respectively. The absorptance of TE-polarized light with incident angles 15°–75° for TiO_2 with thickness $h = 18$, 60, and 86 nm on an Ag substrate is shown in **Figure 2**a–c, respectively, and on an Ni substrate is shown in Figure 2d–f, respectively. Results for TM-polarized light absorption are presented in the Supporting Information.

The strong absorption of the TiO_2-Ag optical coating is limited to wavelengths ≤ 450 nm and its spectral location is independent of the dielectric thickness. This is because Ag is moderately absorptive ≤ 450 nm and thus it is possible to realize strong light absorption even with a lossless dielectric. Using Ni as a substrate, however, allows for strong to perfect light absorption that redshifts as we increase the TiO_2 thickness and spans the visible and NIR regions while maintaining a thickness $h < \lambda/4\, n_D$. The absorption intensity is angular dependent as predicted by Figure 1e,f

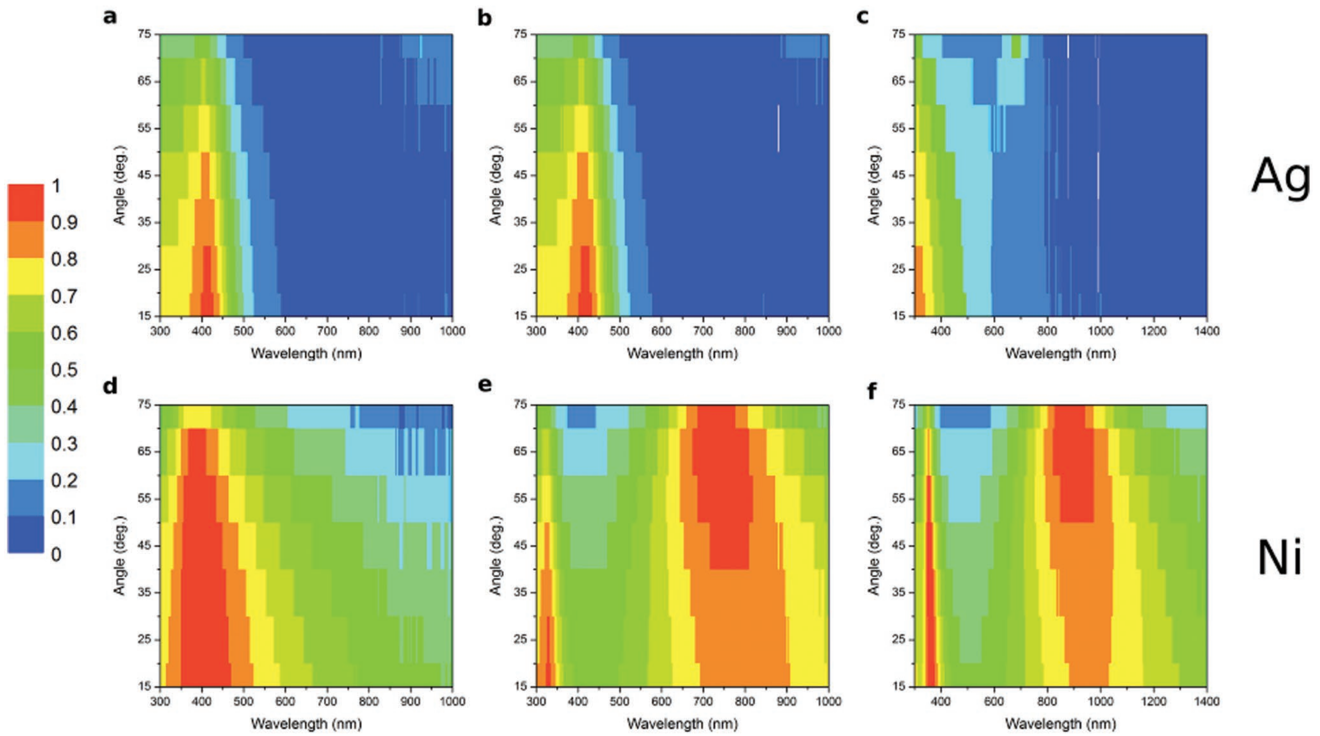

Figure 2. Absorptivity spectra: Experimental absorptivity spectra for TE-polarized light for angles of incidence from 15° to 75° for an Ag film (top) and Ni film (bottom) coated with TiO_2 with thicknesses a,d) 18 nm, b,e) 60 nm, and c,f) 86 nm.

and reaches up to 99.8% for 75° angle of incidence. The ability to control the wavelength and incident angle where SLA takes place highlights the designer properties of the introduced optical absorber. Note that the absorption modes are iridescence free because of the relatively high n_D of TiO_2.[10]

Figure 3a shows an optical image of an Ni substrate coated with different thicknesses of TiO_2 which highlights the possible application of the introduced absorber in structural coloring. Figure 3b shows Ni film deposited on a roughened glass surface (frosted microscope glass slide). The ability to maintain the color integrity even on a roughened surface is due to the iridescent-free absorption. The colors produced are similar to those produced by anodized titanium and other related metals.[19,20] Although oxidized metals are known to show colors, in this work we showed the general principles to realize strong to perfect light absorption for a given dielectric/substrate combination across the visible, near infrared (NIR), and short-wave infrared (SWIR) regimes.

To further demonstrate the importance of the proposed optical absorber, we deposited 75 nm Ge layer on Ag and Ni substrates. **Figure 4** shows the TE-polarization absorptance for incident angles 15°–75°. Since Ge is not highly absorbing above

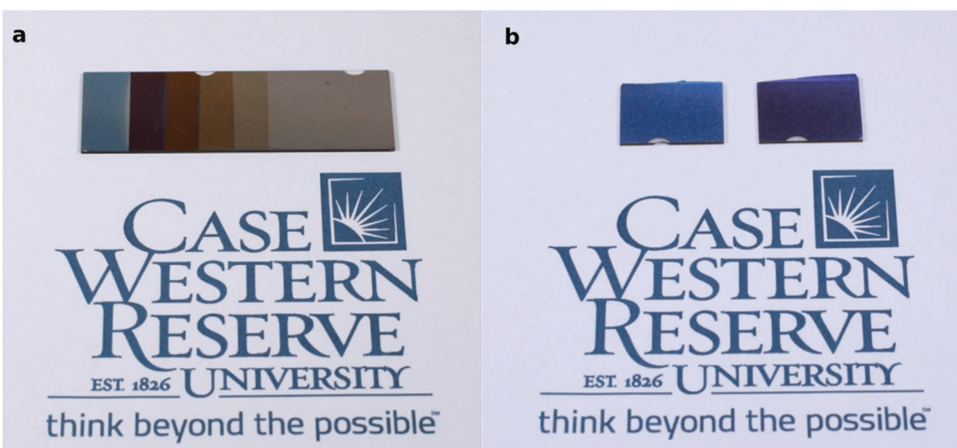

Figure 3. Optical images of optical absorbers: TiO_2 with different thicknesses deposited on an opaque Ni substrate for a) a smooth glass substrate and b) frosted glass substrate. The color integrity on a rough substrate (frosted glass) highlights the practical importance of our absorber for structural coloring.

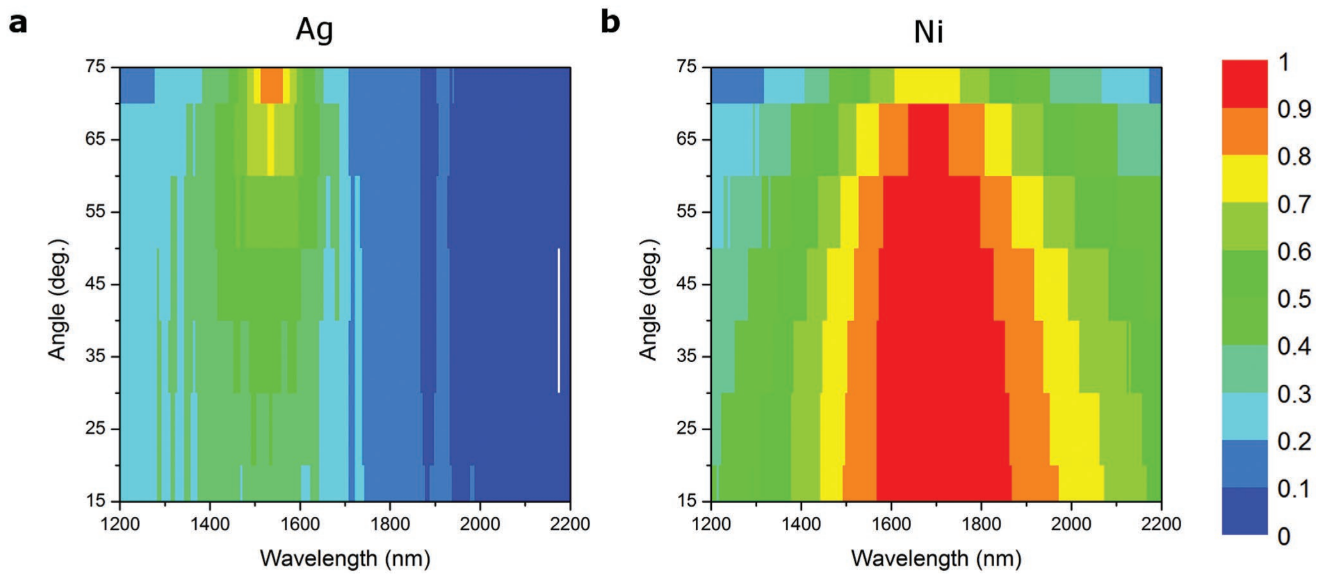

Figure 4. SWIR optical absorber: The absorptivity of an 80 nm thick Ge layer is deposited on a) Ag and b) Ni substrates. SLA is only possible using an Ni substrate since Ge has low losses in the SWIR range.

the SWIR region, strong absorption is realized only for the Ge-Ni structure. In addition, the Ge layer thickness $h \sim \lambda/6\, n_{Ge}$, for $n_{Ge} \approx 4$ at 1750 nm. The absorption reaches up to 99.8% and stays above 90% for all measured angles. Accordingly, we overcame the two main restrictions imposed by the previously introduced optical absorbers, namely, the limited wavelength range where a certain dielectric is absorptive and the limited wavelength range where high reflectivity metals do not behave as a PEC. The introduced absorber has a significant advantage for applications in the infrared regime, for example, as a thermal emitter, especially that metals with low reflectivity, for example, Ti, Ni, and Fe, maintain a nontrivial reflection phase shift for relatively longer wavelengths.[21]

Moreover, the loss mechanism determines the optimal applications for each type of absorbers. We performed a finite-element calculation where we calculated the total power dissipation density of a 650 nm TE-polarized wave incident on two optical absorbers. **Figure 5**a shows the results for an optical absorber based on a lossy dielectric (15 nm Ge) on a highly reflective substrate (100 nm Ag). The losses take place mostly inside the Ge layer, with some losses taking place in the Ag substrate. Figure 5b shows the results for an optical absorber based on a lossless dielectric (40 nm TiO_2) on a moderately absorptive substrate (100 nm Ni). The losses exclusively occur inside the substrate within few tens of nanometers.

The optical absorber based on a lossy dielectric is more advantageous for applications where enhancing the absorption of a semiconductor is important, for example, photodetectors and photovoltaics. On the other hand, for optical absorber based on a lossless dielectric the heat localization in the metallic substrate, which has high thermal conductivity, makes the absorber more suitable for thermo-photovoltaics where the absorbed heat from solar spectrum is transferred to a selective emitter, as well as stealth technology, electromagnetic shielding since high thermal conductivity ensures better heat exchange and management . In addition, the ease of realizing strong light absorption in the IR with dielectric thickness $\ll \lambda$ makes it more suitable for selective thermal emission and radiative cooling. Accordingly, we can engineer the spatial distribution of losses depending on the desired application. Furthermore, it is possible to use the optical absorber

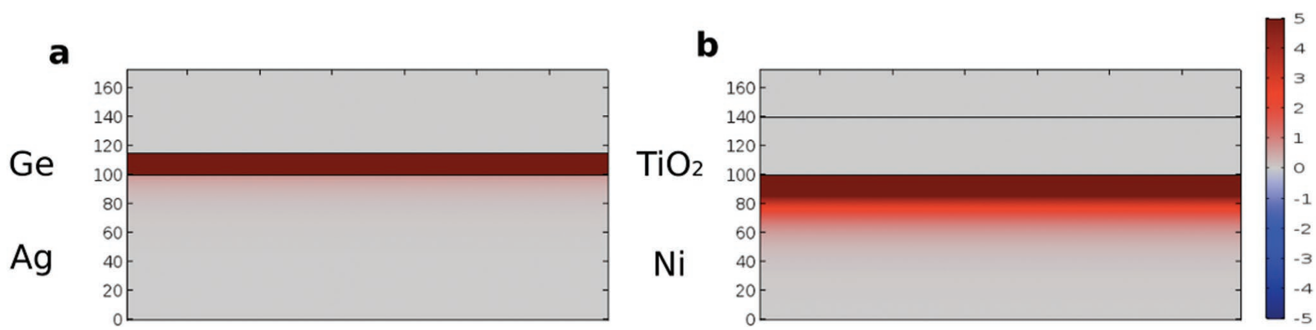

Figure 5. Power dissipation density calculations for TE-polarized 650 nm wave normally incident on a) a 20 nm Ge film on a 100 nm Ag substrate and b) a 40 nm TiO_2 film on a 100 nm Ni substrate.

for gas sensing by using a substrate or a superstrate[22–24] that interacts with a given gas, for example, Pd substrate for hydrogen gas sensing. Adding phase change materials can tune the optical properties and the produced color which is important for data storage and display technologies.[25] Finally, because the optical losses occur in the metal, a heat gradient would exist away from the substrate which can be used for photo-thermoelectric generation.

Experimental Section

Simulations: Finite-element method was used to calculate the power dissipation density in thin films. A commercially available finite element method software was used to calculate the power dissipation density for two thin-film metasurfaces. The first model (Figure 4a) consisted of 20 nm of Ge on a 100 nm Ag substrate, while the second model (Figure 4b) consisted of 40 nm Ni on a 100 nm Ag substrate. A period port was used to excite TE-polarized light at normal incidence to the metasurface, and Floquet periodic boundary conditions were implemented on the left and right boundaries of the simulations. A wavelength domain simulation was used to sweep wavelengths ranging from 400 to 800 nm in steps of 5 nm. In addition, a perfectly matched layer (PML) was placed on the top boundary of the simulation to reduce the effects of reflected light on the top surface.

Thin-Film Deposition: Films were deposited on a glass substrate (Micro slides, Corning) using electron-beam (e-beam) evaporation of Ni, TiO_2, and Ge pellets and thermal evaporation of Ag pellets (both from Kurt J. Lesker). The deposition rate of Ni, TiO_2, Ge, and Ag were 0.5, 1, 0.5, and 10 A s^{-1}. The roughened samples were deposited on a frosted glass substrate (Micro slides, Corning).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

The authors acknowledge support from the Ohio Third Frontier Project "Research Cluster on Surfaces in Advanced Materials (RC-SAM)" at Case Western Reserve University. The authors acknowledge the support of the MORE Center at Case Western Reserve University.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

metasurfaces, optical coatings, perfect absorbers, thin films

Received: May 22, 2018
Revised: August 29, 2018
Published online: October 3, 2018

[1] Z. Zhou, E. Sakr, Y. Sun, P. Bermel, *Nanophotonics* **2016**, *5*, 1.
[2] Y. Pan, G. Tagliabue, H. Eghlidi, C. Höller, S. Dröscher, G. Hong, D. Poulikakos, *Sci. Rep.* **2016**, *6*, 37564.
[3] J. A. Schuller, E. S. Barnard, W. Cai, Y. C. Jun, J. S. White, M. L. Brongersma, *Nat. Mater.* **2010**, *9*, 193.
[4] Y. Yang, K. Kelley, E. Sachet, S. Campione, T. S. Luk, J. P. Maria, M B. Sinclair, I. Brener, *Nat. Photonics* **2017**, *11*, 390.
[5] Z. Li, S. Butun, K. Aydin, *ACS Photonics* **2015**, *2*, 183.
[6] M. ElKabbash, A. Sousa-Castillo, Q. Nguyen, R. Mariño-Fernández, N. Hoffman, M. A. Correa-Duarte, G. Strangi, *Adv. Opt. Mater.* **2017**, *5*, 1700617.
[7] M. A. Kats, F. Capasso, *Laser Photonics Rev.* **2016**, *10*, 699.
[8] M. A. Kats, D. Sharma, J. Lin, P. Genevet, R. Blanchard, Z. Yang, M. M. Qazilbash, D. N. Basov, S. Ramanathan, F. Capasso, *Appl. Phys. Lett.* **2012**, *101*, 221101.
[9] H. Dotan, O. Kfir, E. Sharlin, O. Blank, M. Gross, I. Dumchin, G. Ankonina, A. Rothschild, *Nat. Mater.* **2013**, *12*, 158.
[10] M. ElKabbash, E. Ilker, T. Letsou, N. Hoffman, A. Yaney, M. Hinczewski, G. Strangi, *Opt. Lett.* **2017**, *42*, 3598.
[11] K. Aydin, V. E. Ferry, R. M. Briggs, H. A. Atwater, *Nat. Commun.* **2011**, *2*, 517.
[12] C. M. Watts, X. Liu, W. J. Padilla, *Adv. Mater.* **2012**, *24*, OP98.
[13] J. Chilwell, I. Hodgkinson, *J. Opt. Soc. Am. A* **1984**, *1*, 742.
[14] M. J. Weber, *Handbook of Optical Materials*, CRC Press, Boca Raton, FL **2003**.
[15] J. Simmons, K. Potter, *Optical Materials*, 1st ed., Academic, San Diego, CA **1999**.
[16] M. A. Kats, R. Blanchard, P. Genevet, F. Capasso, *Nat. Mater.* **2013**, *12*, 20.
[17] E. Hecht, *Optics*, 2nd ed., Addison-Wesley, Reading, MA **1987**.
[18] N. Yu, F. Capasso, *Nat. Mater.* **2014**, *13*, 139.
[19] E. Gaul, *J. Chem. Educ.* **1993**, *70*, 176.
[20] M. V. Diamanti, B. Del Curto, M. Pedeferri, *Color Res. Appl.* **2008**, *33*, 221.
[21] M. A. Ordal, L. L. Long, R. J. Bell, S. E. Bell, R. R. Bell, R. W. Alexander Jr., C. A. Ward, *Appl. Opt.* **1983**, *22*, 1099.
[22] C. Wang, L. Yin, L. Zhang, D. Xiang, R. Gao, *Sensors* **2010**, *10*, 2088.
[23] K. V. Sreekanth, S. Sreejith, S. Han, A. Mishra, X. Chen, H. Sun, C. T. Lim, R. Singh, *Nat. Commun.* **2018**, *9*, 369.
[24] K. V. Sreekanth, Q. Ouyang, S. Han, K. T. Yong, R. Singh, *Appl. Phys. Lett.* **2018**, *112*, 161109.
[25] K. V. Sreekanth, H Song, S. Ranjan, *Adv. Mater.* **2018**, *30*, 1706696.