Dual broadband near-infrared perfect absorber based on a hybrid plasmonic-photonic microstructure

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Abstract: High performance light absorber with a broad bandwidth is particularly desirable for near-infrared photodetection and optical interconnects. Here we demonstrate a dual broadband perfect absorber in the near-infrared regime, which is based on a hybrid plasmonic-photonic microstructure. Such a microstructure is fabricated by self-assembling a monolayer colloidal crystal on an optically opaque metal film followed by depositing a thin metallic half-shell on the top of the colloidal particles. Both experimental and numerical simulation results show that the simply designed absorbers have dual broadband with absorption exceeding 90% in the near-infrared region with the absorption bands being scalable by tuning the size of the colloidal particles. Moreover, the absorption efficiency shows an extremely slight dispersion for the incident angles up to 50 degrees, benefit from the high symmetry as well as the highly modulated plasmonic microstructures that lead to a weak polarization dependence of these two absorption bands. The relative ease of growing high-quality colloidal crystals and the low cost of fabricating such plasmonic-photonic microstructures with high reproducibility could promise applicability of the light absorber in the field of photodetectors, thermal emitters and photovoltaics.

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References and links


### 1. Introduction

Due to the coherent oscillations of electrons at the metal-dielectric interfaces, resonant plasmonic nanostructures have been proved to efficiently tail optical responses. Recently,
efficient light absorber based on plasmonic nanostructures has attracted great interest, which could show a wide range of practical applications of photovoltaic cell, optical detector and sensor devices [1–4]. In 2008, Landy et al have proposed an absorbing metamaterial nanostructure consisting of two resonators that couple separately to electric and magnetic fields so as to absorb all incident radiation at microwave frequencies [5]. Near unity absorption was achieved in this structure; however, the designed metamaterial absorbers are polarization-dependent and are only efficiently absorptive over a narrow frequency range. Following this work, many efforts [6–17] have been made to develop new strategies to solve the problem of a narrow bandwidth absorption at a specific resonance frequency since a broadband unity absorption is more desired for many high-performance applications such as the near-infrared photodetection and thermal emitting [18, 19]. One of the most popular and efficient ways to achieve a broad band with unity absorption is to blend different resonators with various strong resonances together. For example, omnidirectional broadband perfect absorbers using multiplexed planar [6–12] or stacking multilayer composite metallic nanostructures [13–16] have been demonstrated. Recently, by engineering the frequency dispersion of metamaterial surface to mimic an ideal absorbing sheet, a broadband infrared absorber has been theoretically proposed by Feng et al [20]. Nevertheless, up to date, the multiband metamaterial absorbers still require structures with more sophisticated geometries in order to induce additional resonances for each absorption enhancement, which introduce additional restrictions on the fabrication process [1].

In contrast with the metamaterial absorbers prepared by utilizing state-of-the-art nanofabrication techniques, a low-cost and easy fabrication method based on the colloidal crystals (CCs) has been employed to fabricate microstructures with resonant light absorption [21–24]. For instance, Teperik et al. have experimentally and theoretically demonstrated that an omnidirectional and polarization-independent absorption effect relying on the excitation of localized plasmons could be achieved using a nanoporous metal surface prepared by electrochemical deposition of gold over CCs supported on a gold substrate [21]; and a perfect absorber consisting of a tungsten films structured by truncated spherical voids array also has been designed to realized a broadband unity absorbance with characteristics of independence of incident angle and polarization in near infrared-visible regime [24]. More recently, a new strategy to dramatically improve light absorption performance has been proposed by forming colloidal spherical dielectric nanoshell resonators that support a low Q-factor whispering-gallery resonant mode [25]. Considering the presence of versatile optical properties including plasmon, photonic and their hybrid modes in the plasmonic-photonic crystals templated using the low-cost colloidal particle arrays [26–30], it is interesting and important to investigate how these controllable optical management approaches can be further extended for achieving a near-unity broadband light absorption in the near-infrared region.

In this paper, we report for the first time a dual broadband wide-angle and polarization-independent perfect absorber in the near-infrared region using a novel hybrid plasmonic-photonic microstructure that supports intrinsic broadband plasmon resonances and multiple hybrid resonant modes [26–30]. Our hybrid plasmonic-photonic microstructures are fabricated by forming a two-dimensional (2D) CC on an optically opaque metal film followed by a coverage of a thin metal layer deposited on top of the 2D CC. We observed a dual broad absorption band in the near-infrared region with bandwidths of about 150 nm and 350 nm respectively, both with a high absorption exceeding 90%. For each of these two absorption bands, the maximum absorption characteristics sustains even for the incident angles up to 50°. Considering the attractive merits of our approach, the present study could provide a guideline to solve the inherent narrow absorption bandwidth and fabrication issues existing in previous plasmonic and metamaterial perfect absorbers [1].
2. Sample preparation and optical characterization

The microstructures under study are formed by first self-assembling a 2D CC of polystyrene (PS) colloids on the surface of a quartz substrate pre-coated with an optically opaque gold film with a fixed thickness \((h)\) of 100 nm, followed by further depositing a thin gold layer with different thickness \((t)\) on top of the 2D CC as shown in Fig. 1(a). A large area of 2D CC can be easily fabricated using a well-developed self-assembly method [31]. As examples, two sizes of the PS colloids of \(1.0 \pm 0.015 \, \mu m\) and \(1.1 \pm 0.017 \, \mu m\) in diameter are used in the present study. Figure 1(b) shows an optical image of the prepared hybrid plasmonic-photonic microstructure based on a highly ordered 2D CC composed of PS microsphere with \(1.1 \, \mu m\) in diameter, which exhibits a uniform color across the whole area of the sample; and a magnified SEM (FEI Philips XL-30) top-view image of a typical area of the microstructure is shown in Fig. 1(c). To characterize this hybrid plasmonic-photonic microstructure clearly, a cross-sectional schematic illustration [Fig. 1(d)] and the corresponding SEM image [Fig. 1(e)] of the sample show the top gold half-shells covered on the PS microspheres with a hexagonal lattice and a thick gold back layer supported on a quartz substrate. Transmission and reflection spectra were obtained using a commercial Fourier-transform infrared (FTIR) spectrometer (Nicolet 5700). The optical spot size of the incident beam impinging on the sample was about 0.8 mm. Polarization-dependent measurements were performed with an adjustable polarizer supplied with FTIR spectrometer. For calibration of angular dependence of reflection, we first measured the reflection spectra under different incident angles from a 100 nm-thick gold mirror as the corresponding baselines, and all the measured reflection from microstructures was then calibrated using these baselines.

3. Results and discussion

Due to the restriction of our equipment setup, transmission \((T)\) and reflection \((R)\) measurements were first performed under an incident angle of \(8^\circ\) with transverse magnetic (TM) polarization [see in Fig. 1(d)]. The absorption \((A)\) was obtained with a definition of \(A = \)
For this hybrid plasmonic-photonic microstructures, a 100 nm-thick gold back plate is exploited to prevent light transmission in the near-infrared regime of interest, which reduces the absorption to $A = 1 - R$. In Fig. 2(a), two broad absorption bands are observed which are centered at 1.15 $\mu$m and 1.85 $\mu$m with maximum absorption values of 97% and 99% for the microstructures consisting of PS colloids with a diameter of 1.1 $\mu$m, covered by gold half-shells with nominal thickness of $t = 9$ nm. The bandwidths with absorption exceeding 90% for the absorption band at shorter and longer wavelength are 150 nm and 300 nm, respectively. It is noted that for a hexagonal lattice with a period of 1.1 $\mu$m, the calculated cutoff wavelength for the onset of 1st order diffraction [21] is located at $\lambda_{\text{cut}} = 0.95$ $\mu$m which is below the short wavelength explored here. Meanwhile, the almost same spectral profile of absorption response is observed in the similar hybrid plasmonic-photonic microstructure when diameter of the PS colloids was reduced to 1.0 $\mu$m [see Fig. 2(a), blue line], and the center wavelengths of the absorption bands are expected to show a blue-shift which is estimated to be in proportion to the size of the PS colloids.

![Fig. 2. Measured (a) and calculated (b) absorption spectra of the hybrid plasmonic-photonic microstructures fabricated using CCs consisting of PS microspheres with diameters of $D = 1.0$ $\mu$m and 1.1 $\mu$m under an incident angle of $8^\circ$. (c) Absorption spectra of the hybrid plasmonic-photonic microstructure (diameter of PS microspheres $D = 1.1$ $\mu$m) under an incident angle of $20^\circ$ as functions of the polarization angle. For all microstructures, the nominal thickness of the Au layer on the top of PS colloids is $t = 9$ nm.](image1)

To understand the unique absorption response, three-dimensional numerical simulations are carried out using a commercial software package (COMSOL Multiphysics) based on finite-element method. The calculation domain constitutes one complete and four-quarter metallic/dielectric core-shells in the xy-plane defined as Fig. 1d. Periodic boundary conditions are applied to the four sides of the rectangular calculation domain to fit the periodicity of the
whole structure [27, 28]. In the simulations, a plane wave with linear polarization (TM) is normally illuminated on the microstructures. The lattice period of the microstructure is equal to the diameter of PS microsphere without regard for the real size distribution of the colloids. The thickness of the flat gold back layer is 100 nm which is larger than the penetration depth of electromagnetic waves in the infrared regime, leading to zero transmission in the working spectral range of our microstructures. The refractive index of the PS \( n_{ps} \) is taken as 1.59 and the permittivity of gold is described by the Drude model \( \varepsilon_{Au} = 1 - \frac{\omega_p^2}{(\omega^2 + i\omega \gamma)} \) with the plasma frequency \( \omega_p = 8.99 \text{ eV} \) and the collision frequency \( \omega_c = 0.0269 \text{ eV} \) [28]. Based on the SEM images in previous reports [31–33], gold distribution on the upper half of PS microsphere would be approximated in a calculated model. However, no theoretical model is yet capable of precisely modeling such a quasi-3D metal microstructures. In order to mimic a relatively thick base of gold half-shell in comparison with the rim area of that, the PS microspheres are assumed to be half-coated by gold half-shells with a semi-ellipsoidal outer shape [33]. In the simulations, the minimized mesh size is set to be 4 nm. Considering the extremely thin gold half-shells in the experiment and the relative large calculating region comparing with very small mesh size in the simulation, the gold half-shells have to be chosen to have a little bit larger thickness of 13 nm on the top of the PS microspheres and 4 nm at the rim in the simulation model, which is expected to yield a good match between experiment and modeling. Figure 2(b) shows the calculated results for the microstructures composed of PS colloids with 1.1 \( \mu \text{m} \) and 1.0 \( \mu \text{m} \) in diameter. Overall, the agreement between the measurements and numerical simulations in terms of the spectral shape is reasonable, considering the fact that the gold distribution on the PS colloidal microspheres might be non-uniform or even not forming a really perfect half-shell [32], making it quite challenging to model the real distribution of gold on the surface of the PS CC. However, based on the above simple model, the numerical simulation shows an obvious frequency shift of the perfect absorption band when different sizes of the dielectric microspheres are exploited, in good agreement with the experimental observations. This proves that the absorption band can be tuned by scaling the dimensions of the dielectric colloids. Actually, the simulated absorption curves do not agree very well with the measured ones, especially for the absorption band at shorter wavelength. A couple of absorption resonances in the higher frequency region instead of a continuous absorption band as we obtained in the experiment are observed. This most likely arises from the relatively simple models [33] adopted in our simulation and the complicated actual distribution of the gold on the surface of the 2D CC.

In order to achieve a polarization-insensitive perfect light absorber, a couple of strategies based on a precise design of the plasmonic or metamaterial microstructures have been developed in previous reports [6–9, 13–15, 21]. Here we show that the dual absorption bands have weak dependence on the polarization angle, particularly for the band located at a short wavelength region. Still for our microstructure composed of 1.1 \( \mu \text{m} \) diameter PS colloids, Fig. 2(c) plots the light absorption spectra as a function of the polarization angle under an incident angle of 20°. Clearly, the absorption bandwidth, center wavelength as well as the absorption amplitude maintain nearly unchanged as the polarization angle is varied from 0° (TM polarization) to 90° (TE polarization) [defined in Fig. 1(d)]. This polarization-insensitive light absorption could be attributed to the spherical morphology of the plasmonic resonant microstructure formed on the top of the dielectric CC as well as the highly symmetric configuration of the hexagonal lattice [21, 26–28].

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Fig. 3. Calculated $|E|^2$ map of the two resonant absorption modes at $\lambda_1 = 1.97 \mu m$ (a,c,d) and $\lambda_2 = 1.34 \mu m$ (e,g,h) for the hybrid plasmonic-photonic microstructures fabricated using CCs of PS microspheres with 1.1 $\mu m$ in diameter. Color map is in linear scale and the same scale is used for all maps. (a) A cross-sectional view at $\lambda_1 = 1.97 \mu m$, and its two top views with $z = 825$ nm (c) and $z = 100$nm (d). (e) The cross-sectional view at $\lambda_2 = 1.34 \mu m$, and its two top views with $z = 825$ nm (g) and $z = 100$nm (h). The white dashed lines mark the $xy$ planes at these two different $z$ values. (b) and (f) Electric field vectors mapped on the $xoz$ plane across the center of one microsphere for $\lambda_1$ and $\lambda_2$, respectively. Arrows represent field directions and colors show strength with red larger and black smaller. The blue dashed circles outline the regions of the gold coated microsphere microstructures. Black signs “+” and “−” stand for positive and negative charges, respectively.

To gain insight on the mechanism of the enhanced absorption, the electric field intensity distributions ($|E|^2$) associated with the observed near-unity absorption bands in our hybrid plasmonic-photonic microstructure are plotted. Figure 3 shows the cross-sectional and top views of the electric field intensity distribution evaluated for the sample containing 2D CCs of PS microspheres with diameter 1.1 $\mu m$. Note that the normalized electric field intensity distributions are mapped using the same scale for the convenience of comparison. The electric field intensity distributions are calculated for the wavelengths $\lambda_1 = 1.97 \mu m$ and $\lambda_2 = 1.34 \mu m$, which are located at the center of each absorption band. Figure 3(a) shows the electric field intensity profile on a cross-sectional plane cut through the center of a PS microsphere (in the $xoz$ plane) for the longer wavelength band centered at $\lambda_1 = 1.97 \mu m$. Also, the electric field intensity distributions in the sectional plane ($xoy$ plane) at $z = 825$ nm and the interface between back gold plate and 2D PS CC at $z = 100$ nm are plotted in Figs. 3(c) and 3(d), respectively. It is found that the electric field intensity is strongly confined on the outer surface of gold half-shells. Such kind of field patterns could be more clearly seen from its vector field plot. As shown in Fig. 3(b), the charge distributions are schematically marked as “±” signs according to the direction of the electric fields closed to the outer surface of the gold coating layer, in which strong electric field is clearly observed. This field pattern is the typical characteristic of a localized plasmon mode supported by a metallic half-shell, which usually has a broad band feature [26–28]. This broadband plasmon resonance is identified as
a superradiant mode originating from the sphere-like plasmon resonances of the individual metal shells [27, 28], which contributes to a broad bandwidth of the absorption of our microstructure, and this intrinsic broad plasmon resonant band is quite different from the inherent narrowband as the result of the impedance match due to the magnetic and electric resonant coupling in the metamaterial light absorber [1].

For the absorption band of shorter wavelength centered at $\lambda_2 = 1.34 \, \mu\text{m}$, besides the strong electric field intensity distribution on the outer surface of the gold half-shells, most of the electric field intensity is found to be trapped in the PS microsphere and the interstice between the PS colloid and the bottom gold layer. As is shown by the electric field intensity profile along the $xoz$ plane in Fig. 3(e), two lobes of strong-field region are observed, which is the typical feature of guided mode (GM) supported in the PS colloid [22, 29, 30]. The GMs of the PS colloids could couple with each other due to the near field interaction, which finally leads to slab dielectric GM-like modes confined in the 2D CC [22, 29, 30]. Meanwhile, strong electric field intensity bounded at the region between the gold layer and the PS colloids is also obvious, which stems from the excitation of the surface plasmon (SP)-like mode supported by the structure [29, 30]. Similarly, the field maps in the $xoy$ plane at the position of $z = 825 \, \text{nm}$ and $100 \, \text{nm}$, are plotted in Figs. 3(g) and 3(h), respectively. The electric field vector pattern is also plotted in Fig. 3(f), in which obvious strong electric field distributions are observed in the PS colloids and the interfaces between the PS colloids and the up gold coating layer and the bottom gold substrate. Generally, the origin of enhanced absorption has a strong correspondence with the quality of field confinement. In this case, due to excitation of the coupled GM-like and plasmon modes and their hybrids as well, near-unity absorbance would take place in the infrared regime centered at $\lambda_2 = 1.34 \, \mu\text{m}$.

![Fig. 4. Absorption spectra of hybrid plasmonic-photonic microstructure with $t = 9 \, \text{nm}$ under TM polarization (a) and TE polarization (b) as a function of incident angle and wavelength.](image)

We conducted additional experiments to investigate the incident angle dependence of the absorption for our hybrid plasmonic-photonic absorber. Figure 4 shows the measured absorption maps as a function of both wavelength and angle of incidence under TM and TE polarizations. The incident angle [marked in Fig. 1(d)] changes from $\theta = 10^\circ$ to $50^\circ$ with an interval of $2^\circ$. It is obvious that these two broad absorption bands could still be achieved even at large incident angles, and the spectral shape and the absorption amplitude are almost preserved in the wavelength region of our interest. Taking the longer wavelength band with
absorption exceeding 80% into account, a large bandwidth of 500 nm shows only a narrowing of 20 nm as θ increases up to 50°. We attribute the above characteristic of absorption band to the localized nature of the plasmon mode supported by gold half-shell on each PS colloid and the high symmetry trait of the hybrid plasmonic-photonic microstructure. It's noticed that, with θ increasing, both absorption bands show a slight angular dispersion, which might be led by the GM-like modes and the Bragg-like plasmon resonance arising from microstructure periodicity which is defined by the 2D PS CC pattern [29, 30]. Overall, the microstructure could keep the performance of very high and broadband absorption in the infrared regime under wide incident angle range.

Fig. 5. Measured absorption spectra of hybrid plasmonic-photonic microstructures with increasing nominal thickness of the corrugated metal layer t from 0 to 12 nm.

Our previous experiment demonstrated the change of optical response, occurring along the increase of the thickness of gold deposited on 2D CC, which actually induces some topological changes of the gold half-shell [31]. In this work, we study the absorption spectra of a series of samples with different nominal gold half-shell thicknesses (t) from 0 nm to 12 nm, which were prepared by increasing the amount of gold deposit on a 2D PS CC patterned on 100-nm-thick gold film supported by a quartz substrate, as shown in Fig. 5. For a sufficiently thin gold layer (t ≤ 4 nm), the deposited gold nanoparticles form isolated aggregates on the upper part of PS microspheres. In such cases, each of the optical absorption spectrum presents very similar to that of the bare monolayer array of PS microsphere on a gold flat bottom layer, and all the sharp absorption peaks are associated with eigen modes, which either guided in the CC slab due to the refractive index contrast or localized mostly at the interface with the metal [29–31]. As gold deposition is increased gradually from 6 nm to 10 nm, an obvious broad absorption band in the longer wavelength region would be observed,
and the absorption is increased up to about 99% ($\lambda = 1.836 \mu m$). During this process, the coated layer on the microsphere array is electrically separated between adjacent microspheres but might be already continuous (gold half-shell) on each PS microsphere [31], which could support a sphere-like plasmon mode, as shown in Fig. 3(a), leading to the above mentioned broadband absorption. Meanwhile, in the short wavelength range, another broadband absorption would be appeared as the formation of a continuous gold half-shell on each PS microsphere, which originates from the SP-like modes supported on gold half-shell and the interface between PS microsphere and gold back plate, GM-like modes in 2D CC slab, and their hybrids as well [29, 30]. As the three absorption spectra with $t = 8$ nm, 10 nm and 12 nm shown in Fig. 5, the dual broad band with near-unity absorption still maintains, which suggests a tolerance of thickness ($t$) for the requirement in further research and applications. When $t$ is further increased from 12 nm up to about 30 nm, the absorption band will be narrowed with an obvious blue-shift, although the maximum absorption efficiency still exceeds 90% (results not shown here). In addition, with increasing $t$, our preliminary measurements indicate that the gold layer covered on the PS CC shows an electric evolution from insulation to conducting and might be with quite different geometry [31]. Nevertheless, the thickness of deposition gold layer can also give us an additional freedom degree to tune the absorption behavior.

4. Conclusion

In summary, we experimentally and numerically demonstrate a dual broadband perfect absorber in the near-infrared regime by utilizing a novel hybrid plasmonic-photonic microstructure, which is prepared by self-assembling a monolayer CC on a flat optically opaque metal film followed by depositing a thin metallic half-shell on the top of the colloidal microspheres. This fabrication technique is very simple, cost-effective, straightforward and highly reproducible. Both experimental and numerical simulation results show that the absorber has two broad absorption bands with absorption exceeding 90% in the near-infrared region. Both of bands show a slight dispersion for the incident angles and extremely weak polarization dependence. It is noted that our study shows that the dual broad absorption bands are supposed to be tuned in a wide frequency region by scaling the colloid's dimensions, even to the visible regime. With its excellent performance, this new system will provide alternative approach to design and study the broadband perfect absorption and its potential applications in photovoltaic and optoelectronic devices.

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