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Large-area, low-cost, ultra-broadband, infrared perfect absorbers by coupled plasmonic-photonic micro-cavities

Guiqiang Liu[a], Yi Liu[a], Xiaoshan Liu[a], Jing Chen[b,c], Guolan Fu[a], Zhengqi Liu[a,*]

[a] Jiangxi Key Laboratory of Nanomaterials and Sensors, Provincial Key Laboratory of Optoelectronic and Telecommunication, College of Physics and Communication Electronics, Jiangxi Normal University, Nanchang 330022, China

[b] College of Electronic Science and Engineering, Nanjing University of Posts and Telecommunications, Nanjing 210046, China

[c] National Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, China

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ABSTRACT

Perfect absorption of electromagnetic wave is emerging as a promising technology for its potential applications in harvesting solar energy, infrared detection and photovoltaic devices. Recent studies have reported a variety of plasmonic or meta-material structures, which are designed and fabricated to provide broadband absorption by utilizing multiple resonators in a composite unit cell. Coupled plasmonic-photonic micro-cavities, formed by using the colloidal crystal that does not have closed packed structure as the structural template, are surprisingly found to be efficient infrared absorption devices in an ultra-broadband frequency range. Taking the absorptivity above 0.9 into account, the operation bandwidth is up to 2.851 μm in the infrared range. The excellent absorption capability can be attributed to the cooperation effects of the plasmonic resonances by the metal caps and the photonic modes by the colloids. The absorber exhibits polarization-independent and wide-angle behaviors. These findings not only reveal the advantages of colloidal crystals as low-cost materials for ultra-broadband absorbers, but also provide inspiration for future development of high-performance, large-area infrared optoelectronic devices.

1. Introduction

Although the electromagnetic wave absorbers have been developed for years since the significant reports in 2008 [1,2], the feasible way to achieve the material with ultra-broadband perfect absorption via a low-cost fabrication technique for large-area structures has been long pursued. In general, light absorption can be obtained via the excited plasmonic resonances by the structured metal surface. Resonant light absorption has been widely studied theoretically and experimentally in the metallic structures such as metallic gratings [3], nano-particles [4] and nano-slits [5]. Meanwhile, based on the structural scheme of the metal-insulator-metal layered structures, meta-materials have been explored to yield high absorption [6–8]. Nevertheless, owing to the intrinsic single resonance of the plasmonic nano-structure and meta-material, these resonant absorption platforms have been designed to absorb light within a narrowband spectrum. As for the potential applications on the broadband thin-film solar energy absorbers and photovoltaic cells [9–13] and anti-reflection devices [14], the method for achieving a resonant absorption band that spans a broad wavelength range is desired.

Since the resonant absorption usually occurs at a certain frequency determined by the geometry of the meta-material, a feasible way to expand the absorption bandwidth can be obtained by using this property. Indeed, the most popular and effective way to extend the absorption bandwidth is proposed and practiced by integrating a series of sub-resonators with different structural sizes into a composite unit cell, which can therefore provide different plasmonic resonant absorption peaks in the spectral range. Moreover, based on the precise design of the resonators and the following absorption bands to overlap each other in the frequency range, broadband and ultra-broadband absorption can be realized. For instance, gradually varied plasmonic resonators with almost continuously changing of the structural size and dimension such as a metallic trapezoidal [15] or metal-insulator layered saw-tooth geometry [16] and structured metal films [17–19] have been theoretically proposed and experimentally demonstrated. However, accompany with the improving of absorption bandwidth, the high-cost and the complex, elaborate fabrication requirements inevitably make them greatly difficult to be produced over large areas and eventually hinder their potential applications.

In contrast to the plasmonic resonances by the metallic nanostructures, optical resonant modes in the photonic dielectric cavities have been demonstrated with strong contribution for the improved

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

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light absorption. The efficiency of the designed solar cell has been numerically observed to be significantly enhanced by coupling the solar irradiation with the whispering gallery modes in micro-spheres [20,21]. Moreover, efficient light trapping in a wide-band spectral range has been experimentally demonstrated via a new approach to form resonant modes by the spherical nanoshell structure [22]. Indeed, two-dimensional dielectric micro-spheres colloidal crystals have been reported with strong resonant optical modes [20–23]. Plasmonic-photonic crystals have also been demonstrated with both plasmonic resonances and photonic modes due to the cooperative effects by the metallic and dielectric resonators [24,25]. As for the colloidal crystals, several impressive advantages can be counted one by one. First of all, the structure can be fabricated via the self-assembled method without the need of high technical processes such as the electron or photo-lithography. Second, the structure can be fabricated in large area. In contrast to the extremely limited sample size scale for the previous reports, this feature undoubtedly shows absolute advantage for mass applications in the optoelectronics. Another main advantage is the low-cost for the structural fabrication together with the low consumption for time and power. However, far less attention is further focused on the light absorption manipulation by using colloidal crystals.

Here, in sharp contrast to the most common approach to broaden the absorption bandwidth via adding the number of sub-resonators in a certain unit cell to produce multiple resonant absorption bands in the plasmonic and meta-materials, we have successfully proposed and demonstrated an ultra-broadband absorber by shrinking the resonator size in the monolayer colloids array. Differing from the limited absorption bandwidth in the closely packed colloids array based absorber, a near-unity absorption with absorptivity above 0.9 within a wide spectral bandwidth of 2.851 μm is achieved in experiment by this novel non-close-packed plasmonic-photonic micro-cavities scheme. The impressive absorption behavior mainly results from the plasmonic resonances of the metallic resonators and the optical resonant modes by the photonic cavities and their hybridized coupling effects. By tuning the structural parameters, it is feasible to manipulate the absorption properties. Moreover, the ultra-broadband absorption can be maintained in the complex polarization and oblique illumination, indicating the absorber platform is applicable in occasions where the polarization and/or incident angle constantly vary (*i.e.* the sun irradiation).

2. Experimental method and numerical calculations

2.1. Sample fabrication

Before the self-assembly fabrication of the colloidal crystals, a 100-nm-thick gold film was deposited on a clean quartz substrate via the magnetron sputtering technique (TRP-450, SKY Corp.). The gold film coated quartz glass was then immersed in the solution of the detergent for 12 h, which could produce efficient hydrophilic surface on the gold film. Then, aqueous suspensions of the PS colloids (Duke Corp.) with 1 wt% was injected to the wedge-shaped cell, which is composed of two contact glass slides holding at a small angle of $\sim 10^{\circ}$ [23]. After the injection of PS colloids solution, these cells were maintained horizontally and kept at room temperature with the surrounding humidity of $\sim 60\%$ for 8 h. Owing to the capillary forces in the built sample cells, the colloidal solution immediately spread on the wedge-shaped cells. A large-area high-quality monolayer of colloidal crystal would be grown onto the top surface of the lower substrate via the convective assemblies by the evaporation of the colloidal solution. The fabricated 2D colloidal crystal samples were etched to a reduced size with a proper inter-particle distance using a plasma etching system (March PX-250) [26]. The operation pressure of 80 mtorr, a radio frequency power of 150 W, an O_2 flow rate of 100 sccm (standard cubic centimeters per minute), and a varied etching time from 50 s to 200 s were applied during the reactive ion etching (RIE) process. After the RIE procedure, a 15-nm-thick gold film was deposited onto the PS colloids by the magnetic sputtering procedure.

2.2. Experimental measurement

Spectral absorptivity is defined as $A = 1 - R - T$, where the R and the T respectively represents the reflection and transmission. In this work, the opaque gold film substrate directly cancels the transmission. Thereby, in experiment, the absorptivity can be obtained when the reflectivity is measured. A customized Fourier Transform Infrared Spectroscopy with an angle-varied reflectivity measurement module was employed to measure the spectral reflection. For each case, the relative reflectivity was obtained by comparing the measured values with that of the pure gold film substrate which was taken as the reference. During the measurement, the optical spot of the illumination light beam onto the sample is about 1 mm. A polarizer within the near-infrared region was used in the polarization-dependent measurements. Structural geometries were characterized by the SEM images (S-3400N).

2.3. Numerical calculations

The optical properties and resonant field distributions were estimated from three dimensional numerical calculations using the finite-difference time-domain method [27]. Periodic boundary conditions were adopted in the x- and y-directions to reproduce the periodic plasmonic-photonic micro-cavities array. Perfectly matched layers were imposed at the boundaries along the z-directions. The relative permittivity of gold was obtained from the experimental data [28] and described by the Drud mode, $\varepsilon_{\mathrm{Au}} = 1 - \omega_{\mathrm{p}}^2/[\omega(\omega + i\omega_{\mathrm{c}})]$, with the plasma frequency $\omega_{\mathrm{p}} = 8.99$ eV (2.171×10^{15} Hz), and the collision frequency $\omega_{\mathrm{c}} = 0.0269$ eV (6.495×10^{12} Hz) [23]. The permittivity of the PS was taken as 1.59. The spectral absorptivity was defined as $A = 1 - R - T$. Because of the use of opaque gold substrate, there was no light transmission through the structure. The absorptivity can be directly achieved by $A = 1 - R$.

3. Results and discussion

Monolayer colloids array can be fabricated in large-area scale via the self-assembly method [23–25,29]. These two-dimensional (2D) colloidal crystals have been widely used for photonic manipulations in nonlinear optics [30], waveguide and filtering [31]. In addition, the monolayer colloidal crystals have also been used as the convenient templates for plasmonic architectures, which have been demonstrated for high-Q plasmonic-photonic hybridized resonances in the metallo-dielectric crystals [24], and even for the broadband transparent conductors [32]. Although various applications were demonstrated in these kinds of devices by using the colloidal crystals, far less attentions have been given for its potential applications in the enhancement of light absorption, and only with few exceptions. For instance, by using the 2D monolayer colloids array as a template, spherical voids arrays buried in metal have been demonstrated with an omnidirectional high absorption, which mainly resulted from the excitation of localized plasmonic resonances in the metallic micro-cavities [2,33]. Based on the emerging of the technique for fabrication non-close-packed colloidal crystals [34], ultra-thin light absorption devices [35] have been developed in these years. Nevertheless, the potential approach for achieving ultra-broadband light perfect absorption by the easily and simply preparative, low-cost, large-area colloidal crystal is still unexplored even though there were several investigations on the high-Q or broadband light absorption by the metallo-dielectric crystals [24] and/or the multiple resonators composite random plasmonic structures [25]. In this work, by combining the nonclose-packed colloidal crystal with the metal films, a spatially separated metal caps-colloids-metal reflector array is proposed, which eventually forms a novel coupled plasmonic-photonic micro-cavities array. The metal caps were the metal half-