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Light-driven tunable dual-band plasmonic absorber using liquid-crystal-coated asymmetric nanodisk array

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We experimentally demonstrated a light-driven reconfigurable near perfect plasmonic absorber working at dual frequencies in infrared range. By employing nanodisks with different sizes in certain arrangement, near perfect absorption of incident electromagnetic waves can be achieved for different working frequencies due to the resonance between the incident light and the nanodisk of different sizes. We showed that optically induced changes in the dielectric constant of the adjacent liquid crystal layer is an effective means to tune the absorption bands of an asymmetric gold nanodisk array. Our liquid crystal based infrared plasmonic absorber can be tuned by using visible light in real time. A tunable range of 25 nm has been confirmed by both simulation and experiment. © 2012 American Institute of Physics. [doi:10.1063/1.3681808]

Plasmonics has been a focus of intensive research for its potential applications in super-resolution imaging,^{1–4} cloak-ing,^{5,6} nanofabrication,^{7,8} energy harvesting,^{9,10} and plasmonic circuitry.^{10–14} Among these applications, plasmonic solar cells have been reported by using nanodisks or nanoparticles to modify the surface profile to allow the confinement of more solar energy inside the functional layer of the solar cell, leading to improved efficiency for a plasmonic solar cell over conventional solar cells.9 Such nanodisk/ nanoparticle pattern can also act as a plasmonic absorber, the so-called plasmonic blackbody^{15,16} as demonstrated by Kravets et al. In particular, Na et al.¹⁷ and Hao et al.¹⁸ used a three-layered plasmonic absorber to achieve a resonance between the top metal structure and bottom metal layer, and selective conversion of light energy into plasmonic resonance leads to almost perfect absorption. These plasmonic absorbers hold great promise for advancing solar cell technology.

Recently, significant amount of research has been focused on developing plasmonic absorbers with extended working frequency ranges or multiple resonances.^{10,19,20} The performance of the plasmonic absorber is determined by its engineered configuration; thus, it is hard to reconfigure the device or tune the absorption bands without redesigning and repeating the entire fabrication process. In this regard, an actively tunable plasmonic absorber could be used to optimize the efficiency of solar cells, which usually exhibit narrow absorption range in near IR and visible. In this article, we demonstrate a real-time reconfigurable dual-band plasmonic absorber by incorporating a photosensitive nematic liquid crystal (PNLC) layer onto an asymmetric gold nanodisk array. The photosensitivity of PNLC arises from the azo-dye dopants, which can change the refractive index of the liquid crystals through several mechanisms.²¹ Compared to other tuning mechanisms such as electric fields²² and acoustic waves,²³ optical tuning is preferable since no electrode or other direct contact is needed.²⁴ In this proof-of-concept study, the refractive index change is attributed to thermal effects caused by light absorption by the azo-dyes. We have observed a tuning range of ~25 nm in the dual absorption bands of the plasmonic nanodisk array.

Figure 1(a) shows a schematic of the near perfect plasmonic absorber. It comprises two different sizes of gold nanodisks, which exhibit two localized surface plasmon resonances (LSPR).²⁵ These resonant frequencies are determined by the size of the metal nanodisks, as well as the inter-spacing between them. As discussed in Refs. 17 and 18, electric or magnetic dipole will form in a three-layer structure. These dipole oscillations will completely convert a specific electromagnetic energy into displacement current between the two metal layers, thereby confining this part of energy inside the layered structure until it completely dissipates through the structure, indicating perfect absorption phenomenon at the specific frequency. Using two different sizes of gold nanodisks (Fig. 1(b)) will therefore result in dual absorption bands.

Figure 1(c) shows the SEM image of a typical near perfect plasmonic absorber we fabricated. Our three-layer structure consists of a specially designed configuration for the top gold pattern to achieve the dual-band near perfect absorption and a 200 nm gold bottom layer to block any incident/transmitted light. The top layer consists of two nanodisk arrays arranged in a two-dimensional (2D) pattern with the same period *d*, as shown in Fig. 1(b). The two nanodisk arrays with different sizes are designed to have different plasmonic resonances interacting with incident light. A thin layer of SiO₂ here (30 nm) is used as the middle spacing layer under the consideration of two criteria: (1) the layer should be thin enough to ensure the sufficient coupling between the top and bottom metal layers, meaning that the plasmonic resonance

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FIG. 1. (Color online) (a) Schematic of light-driven tunable plasmonic absorber. Light-sensitive liquid crystals on top of the absorber can be modulated with light. (b) The top layer consists of two sets of alternatively arranged, two-dimensional nanodisks with the same period and different diameters, followed by a spacing layer of SiO_2 and a bottom gold layer. (c) SEM images of fabricated near perfect plasmonic absorber working at dual frequencies.

in the top layer can reach the bottom layer. (2) A dielectric with relative high dielectric constant is preferred in a capacitor configuration between the top and bottom metal layers. Large capacity leads to enhanced confinement of light energy inside our three-layer structure.

We have performed several numerical modeling and experimental characterization of our plasmonic absorber, and results of these studies are given in Fig. 2. The numerical modeling is performed by using commercial simulation software Lumerical FDTD (Ref. 26) with build-in material parameters (Au and SiO_2) from the material library in the software. The incident electromagnetic wavelengths range is from $0.9 \,\mu\text{m}$ to $2.4 \,\mu\text{m}$ with controllable polarizing angle and incident angle. The periodic boundary condition is applied on both directions of the 2D array. The incident plane is located above the structure and impinging on the structure from the top. An observation plane is positioned above the incident plane to collect the reflected light from the structure. The distance between the observation plane and the structure is 500 nm, which is close enough to collect any scattered electromagnetic waves. Another observation plane is located beneath the substrate to collect any transmitted electromagnetic waves. In our case, there is no transmitted light at all, since, as we mentioned above, the bottom gold layer is thick enough to block all the incident electromagnetic waves. The periods of both large and small nanodisk arrays here are set as 600 nm with thickness of 30 nm for a demonstration case. We provide two different designs with different combinations of large nanodisk and small nanodisk arrays with a = 300 nm, b = 200 nm; and a = 280 nm, b = 180 nm.



FIG. 2. (Color online) (a) Two designs of the near-perfect plasmonic absorber (i) a = 300 nm, b = 200 nm, and (ii) a = 280 nm, b = 180 nm. FDTD simulations show that near perfect absorption can be achieved at selected wavelength ranges. (b) Experiment results of the two designs. Over 90% absorbing efficiencies can be achieved.

Figure 2(a) depicts the corresponding simulated results. Near perfect absorption of the incident electromagnetic waves are represented as two near-zero reflection minima. By resizing two nanodisk arrays, large shifts in these absorption dips can be observed. Experimental confirmation using Fourier transform infrared (FTIR) spectrometer and imaging microscope (Bruker FTIR, HYPERION 3000) is plotted in Fig. 2(b), with the exact parameters used in Fig. 2(a). The results match almost perfectly in terms of the position of the absorption peaks, with slightly reduced absorption when compared to the ideal case in simulations. The slight differences between the experimental and computational results are likely caused by imperfections in the fabrication and different boundary conditions in simulations and real cases. The boundary conditions in the simulation are infinite in a 2D plane (e.g., periodic boundary conditions for large-area nanodisk array); whereas, the measuring area in the experiment is finitely limited by the field of view in FTIR spectrometer. Nevertheless, the measured value of over 90% absorption is arguably near-perfect.

As described in the previous section, the absorption bands of plasmonic absorber can be tuned by resizing the nano-disks through multiple fabrication processes; a more convenient approach is to incorporate a tuning mechanism into the structure. Among the various methods and media^{27–34} that have been used to tune plasmonic resonances, nematic liquid crystals are attractive due to their large birefringence and flexible tuning mechanisms.^{21,35–39} Among those tuning mechanism, optical tuning is most preferred as it does not require electrodes or other direct contacts with the structures. Furthermore, all-optical modulations of the liquid crystal birefringence can be implemented at speeds several orders of magnitude faster than with electro- or acousto-optical approaches. For example, thermal and density changes induced by short-pulsed lasers produce the required refractive index changes in microseconds to nanoseconds.^{35–37}

In this proof-of-concept study, we employ laser-induced birefringence change [Δn] in azo-dye doped nematic liquid crystal. The active medium consists of the liquid crystal 5CB (pentyl-cyano-biphenyl) doped with ~4% of methyl-red dye dispersed onto the three-layered structure. Subsequent probing of the reflection property of the structure with s- and p-polarized light shows very small variation, indicating that the director axis of the liquid crystals are aligned homogeneously on the structure with director axis randomly distributed on the gold surfaces, i.e., the effective refractive index "seen" by the probing light is the isotropic value $n_{iso} \sim 1.61$ [Note: this value is for visible wavelength $\lambda = 0.5 \,\mu$ m, where $n_e = 1.742$ and $n_o = 1.547$; the refractive indices are lower in the near infrared regime $\lambda = 1 \,\mu$ m,⁴⁰ where $n_e \sim 1.63$ and n_o is ~ 1.50 and $n_{iso} \sim 1.54$].

The experimental setup for optical tuning of the absorption dip is shown in Fig. 3(a), with an infrared microscope connecting to FTIR to collect the reflection spectrum at room temperature of 22 °C. An argon laser ($\lambda = 514$ nm) is used here as a pumping laser with its power tunable from 0 to 248 mW. The laser impinges obliquely on the sample so that its reflections are not collected by the microscope objective. When illuminated by the pumping light, several mechanisms could occur in the methyl-red doped nematic liquid crystal.²¹ The most likely mechanism at work here is optically induced alignment of the methyl-red molecules [and therefore the liquid crystals molecules] towards the normal to the gold surface, i.e., the probe will "see" a lower refractive index value closer to the ordinary index value n_0 .^{41–43}

Fig. 3(b) shows the measurement of a sample with a = 300 nm and b = 200 nm. Note that with the liquid crystal overlayer, the two absorption dips are shifted by a considerable amount (from $1.1 \,\mu\text{m}$ (in air) to $1.35 \,\mu\text{m}$, and from $1.52 \,\mu\text{m}$ (in air) to $1.7 \,\mu\text{m}$, respectively). Because of the large index mismatch between the liquid crystal $(n_{iso} \sim 1.61)$ and air (n = 1.0), the minima of the reflection bands are higher. When the refractive index of the liquid crystal is changed by an amount $\Delta n = -0.06$ [from 1.61 to 1.56], the simulation shows that a shift of around 30 nm. The corresponding experiment results are plotted in Fig. 3(c). We can clearly observe the change on absorption dips before and after the pumping light was applied, and the tuning is reversible and repeatable; in our experiment with low cw laser power illumination, the tuning speed is ~ 1 s. We also observe that the performance of plasmonic absorber has somewhat been compromised after introducing the liquid crystal and dye mix-



FIG. 3. (Color online) (a) Experimental setup for measuring optically tunable plasmonic absorber. (b) FDTD-simulated results of the performance of the Liquid crystal-plasmonic absorber. Around 30 nm shift can be observed. (c) Measured absorption band shifts with the pumping light turning ON and OFF. (d) Zoom-in view of the left absorption dip. The shift is around 25 nm. (e) Zoom-in view of the second absorption dip. The shift is around 20 nm.

ture, with one absorption dip giving \sim 70% absorption compared to over 90% before the introduction of liquid crystals. Figures 3(d) and 3(e) show a magnification of the two

resonances from Fig. 3(c); the shifts are around 20-25 nm, which is in good agreement to the simulation results [shift of 30 nm]. By optimizing the geometry, or dye-concentration and utilizing other mechanism or liquid crystals to produce larger birefringence change, one could expect larger and faster tuning for the absorption dips.

In conclusion, we have designed and constructed a lightdriven reconfigurable dual-band plasmonic absorber comprised of an asymmetrical gold nanodisk array functionalized with a layer of photosensitive dye-doped liquid crystals. The tuning process is reversible and repeatable. With further optimization, much larger tuning range and absorption efficiencies, and faster tuning can be attained.

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