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Wide-angle, polarization-independent ultrathin broadband visible absorbers

Kyu-Tae Lee, a) Chengang Ji, a) and L. Jay Guo b)
Department of Electrical Engineering and Computer Science, The University of Michigan, Ann Arbor, Michigan 48109, USA

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A capability of absorbing a broad range of visible lights is essential to boost the performance of various applications, such as photovoltaics (PV), photodetectors, and thermal emitters. Here, we present an angle-insensitive, polarization-independent ultrathin (<150 nm) broadband absorber in the visible regime exploiting strong interference behaviors in highly absorbing semiconductor materials. A proposed structure simply has four layers composed of two stacks of a metal and a semiconductor demonstrating a remarkably enhanced absorption property as compared with the device without a top semiconductor film. This is attributed to multi-cavity resonance effects in each cavity, which is obviously elucidated with phase calculations and electric field distributions. The maximum absorption efficiency of the device is 95.5% at a resonance and its absorption characteristic can be maintained over a wide angle of incidence up to ±70° regardless of the incident light polarization. Finally, we investigate how our approach can be utilized to achieve a tandem PV cell with high efficiency. Our strategy can be applied to other material systems and can be useful in diverse applications, including thermal emitters and PV.

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Light absorption properties in plasmonic nanostructures and metamaterials have attracted substantial attention for their potential in a wide variety of areas such as color filters, a) b) photodetectors, c) d) photovoltaics (PV), e)–f) sensors, g)–h) bolometers, i) optical lithography, j) and thermal emitters. k)–l) Diverse approaches have been proposed and studied to achieve enhanced absorption characteristics in multilayer architectures, m)–n) metamaterials, o)–p) and nanostructures with subwavelength gratings. q)–r) s)–t) Most of the previous works on light absorbers, however, rely on the surface plasmon resonance (SPR) at the interface between a metal and a dielectric, which results in the absorption only within a very narrow wavelength region. This limits many applications, including PV and thermal emitters, both of which require a large absorption over a wide range of wavelengths. In addition to the narrow bandwidth, such SPR-based properties depend strongly on not only the angle of incidence but also a polarization state of incident light, consequently further restricting their potential within narrow limits. An enormous amount of effort has been dedicated for improving the angle-dependent performance by employing a magnetic resonance or a localized SPR (LSPR), particularly in structures consisting of metal-insulator-metal. v)–w) To overcome the polarization-dependent property, two dimensional patterns, such as square patches, x)–y) nanodisks, z)–aa) circular hole arrays, ab) and crossed trapezoid arrays, ac)–ad) are utilized. Even though angle- and polarization-dependent characteristics could be improved with the above strategies, the vast majority of designed structures involve complicated and expensive fabricating processes to create features at the subwavelength scale, thus making them difficult for the applications over a large area. Therefore, there is a critical need to develop a new device architecture that provides improved performance combined with the simplicity.

Here, we propose and experimentally demonstrate an ultrathin (<150 nm) broadband absorber capable of absorbing the light from 400 nm to 650 nm based on multi-cavity resonances in highly absorbing media. The proposed structure comprises two stacks of a highly absorbing material and a metal showing 95.5% absorption at a resonance. Additionally, our device exhibits high performance of the absorption that is insensitive to the angle of incidence up to ±70° as well as the polarization state of incident light. Besides of these features, only deposition method is involved to fabricate our designed absorber, thus providing a powerful scheme for achieving highly efficient device with relatively simple and cost-effective means for large scale that are distinctly different from those observed in the previous reports, involving expensive fabrication techniques such as e-beam lithography. Finally, we discuss the applicability of our proposed scheme to tandem PV technologies for high efficiency PV cells. The strategy described here could have the potential for many applications, such as metamaterials, thermal emitters, and PV.

A schematic view of the proposed visible absorber utilizing the multi-cavity resonance behaviors in ultrathin semiconductor layers featuring angle-insensitive and polarization-independent broadband absorption property is depicted in Figure 1(a). The device structure simply consists of two stacks of a metal and a semiconductor with its optimized thickness to achieve a broadband absorption characteristic with a high efficiency in the visible range. Amorphous silicon (a-Si) material is utilized as a highly absorbing medium due to its strong absorption property at visible frequencies, and silver (Ag) functions as a reflective mirror since it is highly reflective and has the lowest absorption loss among noble metals. Although the overall absorption could be even enhanced by employing lossy

a)K.-T. Lee and C. Ji contributed equally to this work.

b)Author to whom correspondence should be addressed. Electronic mail: guo@umich.edu.
metals such as aluminum (Al), copper (Cu), and chromium (Cr), we aim at designing the structure with the improved absorption property only in the semiconductor layer, thus potentially extending the range of possible applications, including tandem PV systems. More detailed investigations on how the metal and the semiconductor layers affect the absorption behavior of the structure are given in Figure S1 (see supplementary material39). In our absorber design, the thickness of a top metallic film is designed to be optically transparent (30 nm) so that it allows incident light to pass through a middle semiconductor layer to create an additional resonance at a different wavelength, thereby making the bandwidth broad, whereas a bottom metallic layer is 80 nm thick enough to prevent any transmitted light, thus validating the equation \( A = 1 - R \), where \( A \) is the total absorption and \( R \) represents the reflection. In Figure 1(b), a simulated absorption spectrum of the proposed four-layer device (red solid line) is described along with that of the three-layer device (blue solid line) without a top semiconductor film (i.e., metal-semiconductor-metal (MSM)) for the comparison. As can be seen from the figure, our proposed device structure exhibits a much broad absorption performance with the higher efficiency (>95%), which arises from two distinctive resonances appearing at 490 nm and 575 nm, while the MSM structure shows only one resonance peak with a lower absorption efficiency (61.5%) at 550 nm. Although the broadband absorption can also be achieved by a single thick a-Si layer, the charge recombination will be more dominant when the thick structure is applied for solar cells as the diffusion length of a-Si is only tens of nanometers. As a result, the absorption spectra, the net phase shift, which involves a propagation phase accumulation within the semiconductor layer, and the absorption coefficient of the a-Si material at shorter wavelengths also contributes to the broad resonance at 490 nm. The Ag and the a-Si materials are deposited by a thermal evaporation and a plasma-enhanced chemical vapor deposition (PECVD), respectively. This suggests that the design principle discussed here could be easily scalable to large area applications since it only involves the deposition without any patterning process.

To investigate the effect of the multiple resonances on the absorption spectra, the net phase shift, which involves a phase shift occurring upon the reflection from the interface and the propagation phase accumulation within the semiconductor films, is calculated as presented in Figure 3(a). The propagation phase accumulation can be simply obtained with the equation \( (2\pi/\lambda) \cdot 2nd \cos \theta \), where \( \lambda \) is the incident wavelength, \( n \) is the real part of the refractive index, \( d \) is the thickness of semiconductor layer, and \( \theta \) is the direction of wave propagation and is zero for the normal incidence, while the phase shift occurring upon the reflection from each surface can be calculated from the reflection coefficient for transverse electric (TE) (transverse magnetic (TM)) polarization, which is \( r = \frac{r_{12} + r_{23} e^{i2\theta}}{1 + r_{12} r_{23} e^{i2\theta}} \), where \( r_{ab} = \frac{n_a \cos (\theta_b) - n_b \cos (\theta_a)}{n_a \cos (\theta_b) + n_b \cos (\theta_a)} \) and \( (2\pi) \cdot n_1 \cos (\theta) \). It is noted that the net phase shift is divided by \( 2\pi \), so it represents the number of cavity modes. The resonance modes in the top a-
becomes thinner, the resonances move farther apart and the separation between the top Ag film and the a-Si layer increases. The multiple resonances get shifted and the wavelength is shown, given fixed thickness of the optical absorption as a function of the top Ag film thickness (i.e., 4.71 nm). The large absorption coefficient of the a-Si material at shorter wavelengths (i.e., 4.71 nm) is relatively lower than that at 591 nm, the absorption performance at 496 nm could be still high due to the large absorption coefficient with the a-Si material. It is shown that the electric field intensity at 591 nm is relatively lower than that at 496 nm, thus leading to a high Q-factor (i.e., narrow bandwidth with high efficiency). Similarly, the influence of the top a-Si thickness on both the absorption efficiency and bandwidth vary with the top a-Si-Ag stack thickness and the highest absorption efficiency is achieved when the top Ag and the top a-Si layers are designed to be 30 nm and 12 nm, respectively.

Next, we examine the dependence of the absorption efficiency on the thickness change in both Ag and the top a-Si layers. In Figure 4(a), a calculated 2D contour plot of the optical absorption as a function of the top Ag film thickness and the wavelength is shown, given fixed thickness of the top a-Si layer as 12 nm. The multiple resonances get shifted as the thickness of the top Ag layer varies, which is a direct consequence of varied reflection phase shifts at the interface between the top Ag film and the a-Si layer. When the top Ag becomes thinner, the resonances move farther apart and finally merge into a single resonance in the case that the Ag is so thin that the separation between the a-Si layers cannot be perceived by the propagating light. As the thickness of the top Ag film increases, the intensity of the reflected light gets lower, thus leading to a high Q-factor (i.e., narrow bandwidth with high efficiency). Similarly, the influence of the top a-Si thickness on both the absorption efficiency and bandwidth is also investigated with the top Ag fixed at 30 nm as shown in Figure 4(b). The multiple resonances are separated when reducing the thickness of the top a-Si film as the resonance is created at shorter wavelengths with decreased cavity layer thickness. In contrast, with increased thickness of the top a-Si layer, the multiple absorption peaks get closer since the resonance inside the top a-Si film moves toward the longer wavelength range, where the resonance of the middle cavity exists as analyzed above. From this investigation, it is found that the optimal thicknesses for the top Ag and the top a-Si are 30 nm and 12 nm, respectively, in order to achieve the highest absorption performance.

Simulated and measured angular dependences for both TE and TM polarizations are shown in Figures 5(a)–5(d), respectively. The resulting angle resolved absorption spectra and (b) TM and (c) and (d) TE polarizations up to 70°. The angle insensitivity of the proposed broadband absorber is enabled by the negligible propagation phase accumulation within the ultrathin a-Si layers, showing a flat dispersion curve.
of the fabricated device are measured by the spectroscopic ellipsometer (M-2000, J.A. Woollam) at the angle of incidence ranging from 45° to 70° exhibiting good agreement with the simulation. A highly efficient absorption characteristic encompassing the wavelengths from 400 nm to 650 nm is accomplished over a wide range of incident angles up to ±70°. Note that the bandwidth of the optical absorption of our proposed absorber is limited to some extent as it is difficult for the a-Si material to capture wavelengths beyond 650 nm due to its band gap. We also note that the range of the absorption could be broader by utilizing either low band gap semiconductors or lossy metals. The angle insensitive performance could be explained by the fact that the phase shift accumulated during the propagation through the ultrathin a-Si layer is almost insignificant because the a-Si film thickness is much thinner than the wavelength of incident light.\(^{31,32}\) Such an angle robust functionality is greatly crucial for a wide variety of applications, such as PV, photo-detectors, and thermal emitters.

Finally, we explore the possibility of applying our strategy to tandem PV cells. Figure 6(a) depicts the schematic diagram of the tandem-mimicking ultrathin a-Si PV that can be realized by placing our proposed broadband absorber structure onto the indium tin oxide (ITO)-glass. It should be noted that efficient hole and electron transporting layers, such as vanadium pentoxide (V_2O_5) and indene-C_60 bisad-duct (ICBA), need to be inserted for this structure to operate as the PV device with a better band alignment as discussed in the previous works.\(^{31,32}\) In Figure 6(b), simulated absorption spectra in top (black) and middle (red) a-Si photoactive layer are shown. When the device is illuminated from the ITO-glass side, the top cell (#1, a-Si = 13 nm) mainly harvests the shorter wavelengths (i.e., blue ranges), while the middle cell (#2, a-Si = 31 nm) strongly absorbs the green spectral regions, thereby spanning a broad range of visible wavelengths toward high power conversion efficiency. We note that the absorption beyond 650 nm is limited by the a-Si material. The calculated short-circuit current density ($J_{sc}$) values in each cell are found to be 5.62 mA/cm\(^2\) (#1) and 5.64 mA/cm\(^2\) (#2), respectively, using the following equation:

$$J_{sc} = \int_{400nm}^{800nm} \frac{e\lambda}{hc} QE(\lambda) I_{AM1.5}(\lambda) d\lambda,$$

where $e$, $h$, $\lambda$, and $c$ are elementary charge, Plank constant, wavelength, and speed of light, respectively. $I_{AM1.5}(\lambda)$ is AM 1.5G solar radiation spectrum. We assume that $QE(\lambda)$ is equal to the optical absorption spectrum in the photoactive layer (i.e., internal quantum efficiency is 100%). Since the photoactive layer whose thickness is much thinner than a carrier diffusion length of the a-Si material is used in our design, 100% of internal quantum efficiency would be good approximation due to a nearly negligible charge carrier recombination.\(^{31,32}\) Figure 6(c) exhibits the calculated absorption spectra in the photoactive layer of our proposed tandem PV cell together with the single layer based PV device with the same active layer thickness (total a-Si = 44 nm). It is clear that the spectrum broadening mechanism with the multiple

FIG. 6. (a) A schematic representation of the proposed tandem-mimicking ultrathin a-Si PV device, which could be made on ITO-glass. (b) Simulated absorption spectra in both top (black) and middle (red) a-Si photoactive layers along with the calculated corresponding $J_{sc}$ values. The top cell (#1), which has the thinner thickness than the middle cell (#2), primarily captures the shorter wavelengths of solar radiation, while #2 harvests the green portion of solar energy, thus being able to achieve the broadband absorptions for high efficiency. Note that it is difficult for the a-Si material to absorb the longer wavelengths beyond 650 nm due to its band gap. (c) Simulated absorption spectra in the photoactive layer of the tandem-mimicking a-Si PV cell and the single a-Si layer based PV, both of which have the same total photoactive layer thickness, clearly exhibiting that our proposed tandem approach can significantly boost the absorption in the photoactive layer, thereby leading to higher power conversion efficiency. $J_{sc}$ is calculated by assuming that the internal quantum efficiency is 100% and both FF and $V_{oc}$ are taken from our previous work to estimate the efficiency.
resonances allows the absorption property of the tandem PV device to be significantly enhanced, which can result in a power conversion efficiency ($\eta$) of 4.50%, more than twice higher than the $\eta$ of the PV cell with the single photoactive layer (2.12%). These $\eta$ are estimated with the assumed 64% filling factor (FF) and open circuit voltages ($V_{oc}$) 0.60 V and 0.65 V, all of which were taken from our previous work.\textsuperscript{32}

In summary, we have shown a wide-angle, polarization-independent ultrathin broadband absorber in the visible range utilizing the multi-cavity resonance effects in the semiconductor layers. The designed structure is simply composed of four layers (2 stacks of the semiconductor and the metal) and any complex fabrication process (e.g., lithography and etching) is not required to fabricate the device. Putting additional cavity layer with a proper thickness on top of the MSM architecture enables the absorption property to be remarkably enhanced. The absorption peak of 95.5% is achieved at a resonance, which can be retained over a large angular range ($\pm 70^\circ$) under both TE and TM-polarized light illumination. These features are highly desired for various applications such as PV, thermal emitters, photodetectors, and metamaterials.

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39. See supplementary material at http://dx.doi.org/10.1063/1.4939969 for investigations of the dependency of the absorption on various materials and structures. In supplementary Figure S1, we investigate how the metal and the semiconductor layers affect the absorption behavior of the semiconductor-metal-semiconductor-metal (SMSM) structure. In Figure S3, we compare the absorption behavior of the a-Si/Ag/a-Si/Ag stacks and a thick single layer of a-Si. Figure S4 shows even broader and higher efficiency absorption can be achieved with more stacks of a-Si/Ag. The refractive indices of a-Si and Ag used in the simulation are also included in Figure S2.