Zeroth order Fabry-Perot resonance enabled ultra-thin perfect light absorber using percolation aluminum and silicon nanofilms

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Abstract: We demonstrated perfect light absorption in optical nanocavities made of ultra-thin percolation aluminum and silicon films deposited on an aluminum surface. The total layer thickness of the aluminum and silicon films is one order of magnitude less than perfect absorption wavelength in the visible spectral range. The ratio of silicon cavity layer thickness to perfect absorption wavelength decreases as wavelength decreases due to the increased phase delays at silicon-aluminum boundaries at shorter wavelengths. It is explained that perfect light absorption is due to critical coupling of incident wave to the fundamental Fabry-Perot resonance mode of the structure where the round trip phase delay is zero. Simulations were performed and the results agree well with the measurement results.

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References and links

1. Introduction

Enhanced light absorption in nanostructure materials is significant for many applications such as solar energy harvesting, nonlinear optics, and biosensing [1–3]. In the past decade, perfect light absorption in patterned metal film structures [4–9] and un-patterned metal-dielectric films [10–26] has been extensively investigated. Among these perfect light absorption structures, patterned metal structures require complicated nanofabrication processes, whereas un-patterned thin film structures involve only thin film deposition, which intrinsically have low manufacturing cost. Recently, perfect light absorption in single layer silicon (Si) films deposited on an aluminum surface has been reported [24]. The thickness of the silicon film is about 1/5 of the perfect absorption wavelength. In this work, significant reduction in overall layer thickness is demonstrated by adding an ultra-thin Al nanolayer on top of the silicon film. Although metal-dielectric film nanocavity perfect light absorbers have been previously investigated [16–19], here we show that by using silicon film as the cavity medium together with an ultra-thin percolation aluminum film on the top, the cavity thickness can be significantly reduced relative to wavelength for perfect light absorption. It was reported recently [21] that silicon was used as a high index cavity medium for enhanced light absorption in metal-dielectric nanocavity structure, but perfect light absorption was not achieved. In this work, we also found that the ratio of silicon cavity layer thickness to the perfect absorption wavelength decreases as the cavity layer thickness decreases due to increased phase delays (smaller phase gains) at the silicon-aluminum boundaries at shorter wavelengths than at longer wavelengths. Optical phase analysis reveals that perfect light absorption occurs at the fundamental Fabry-Perot resonance mode where the round-trip phase delay in the cavity is zero.

2. Fabrication and characterization of ultra-thin perfect light absorbers

Figure 1(a) shows the schematic of the metal-dielectric film nanocavity structure in which a thin silicon (Si) layer is sandwiched between an ultra-thin aluminum layer on top and a thick aluminum (Al) film on bottom. The bottom aluminum film is thick enough to prevent light
penetration to substrate. This structure is also known as Salisbury screen structure [27], in which wavelength selective perfect light absorption can happen at critical coupling condition.

For device fabrication, a thick aluminum film of 100 nm was first sputtered on a Pyrex glass wafer by using a Denton sputter with 200 W DC power, 5 mTorr argon plasma pressure, 120 seconds pre-deposition conditioning time, and the deposition time of 625 seconds. The deposition rate of Al film was 0.16 nm per second, calibrated by using a surface profilometer (KLA-Tencor P-10). After deposition of the thick aluminum layer, the sample was diced into four pieces. The diced four pieces were used as substrates for subsequent deposition of Si films of 30, 40, 50, and 60 nm thick. For Si film depositions, the Denton sputter was set with RF power of 200 W, argon plasma pressure of 5 mTorr, and a pre-deposition conditioning time of 180 seconds. The deposition rate of Si was 0.1 nm per second. The deposition time of silicon film was from 300 to 600 seconds with the increment of 100 seconds for giving different thicknesses. The 40-nm-thick Si sample was further cut into six pieces for subsequent depositions of ultra-thin Al layer on top. The top Al layer thickness varies from 5 nm to 10 nm with 1 nm increment. After ultra-thin aluminum layers were deposited, optical reflectivity from the sample was measured by using a broadband halogen light source and an optical spectrometer (StellarNet C-SR-50). Figure 1(b) shows the measured optical reflectivity spectra from six devices with different top Al layer thicknesses but same silicon cavity layer thickness of 40 nm. It can be seen that the optical reflectivity minimum first decreases as the top Al layer thickness increases from 5 nm and reaches zero when the aluminum layer is 7 nm. For 40-nm-thick Si device with 7 nm Al layer on top, perfect light absorption occurs at 640 nm wavelength. Further increasing Al layer thickness increases the reflection minimum. A scanning electron microscope (SEM) picture of the 7 nm Al film surface is shown in Fig. 1(b) as an inset. The 7 nm Al film is quasi-continuous with small voids. It is seen in Fig. 1(b) that perfect light absorption occurs for the device with 7 nm Al layer on top, indicating critical coupling of the incident light to an optical cavity resonance mode.

To change the perfect absorption wavelength, we change the thickness of Si layer by keeping the top Al layer constant at 7 nm. Figure 1(c) shows photographs of four fabricated devices with Si layer thicknesses of 30, 40, 50, and 60 nm. The four devices exhibit colors of purple, blue, cyan, and green, corresponding to Si layer thicknesses of 30, 40, 50, and 60 nm, respectively. The reflectivity spectra were measured at 10 degrees angle of incidence. The measurement results are shown in Fig. 1(d). The perfect absorption wavelengths are 540, 640, 750, and 870 nm for Si layer thicknesses of 30, 40, 50 and 60 nm, respectively.

Table 1 summarizes the perfect light absorption wavelengths and corresponding absorbance, as well as the ratio of the silicon layer thickness to the perfect absorption wavelength. It can be seen that the perfect light absorption was achieved over a wide range in the visible and near-IR region while sweeping the silicon cavity thickness from 30 nm to 60 nm. For the perfect light absorber with 30 nm-thick Si film, the ratio of peak absorption wavelength to the silicon cavity thickness is approximately 18. As the Si cavity thickness decreases, the perfect absorption wavelength also decreases. However, Si cavity layer thickness decreases faster than the perfect absorption wavelength decreases. This is because more phase delays (less phase gains) occur in the silicon-aluminum boundaries at shorter wavelengths than at longer wavelengths. Table 1 lists calculated skin depth in aluminum and phase changes in the silicon-aluminum boundaries at different absorption wavelengths. The skin depth of aluminum remains almost constant and does not change much as wavelength changes. This results in more phase delays at shorter wavelengths than at longer wavelengths.
Fig. 1. (a) The schematic ultra-thin nanocavity perfect light absorber structure. (b) Measured optical reflectivity spectra from devices with 40 nm Si thickness and different top Al layer thicknesses from 5 nm to 10 nm. Perfect absorption is observed for the device with 7 nm Al layer thickness. A SEM image of the 7 nm Al film surface is shown as the inset. (c) Photographs of the four devices with different Si layer thicknesses, taken under ambient light. (d) Measured optical reflectivity spectra of perfect absorbers of different Si thicknesses at the near normal angle of incidence of 10 degrees.

<table>
<thead>
<tr>
<th>Silicon cavity layer thickness (nm)</th>
<th>30</th>
<th>40</th>
<th>50</th>
<th>60</th>
</tr>
</thead>
<tbody>
<tr>
<td>Perfect absorption wavelength (nm)</td>
<td>540</td>
<td>640</td>
<td>750</td>
<td>870</td>
</tr>
<tr>
<td>Peak absorbance (%)</td>
<td>99.5</td>
<td>99.9</td>
<td>99.9</td>
<td>100</td>
</tr>
<tr>
<td>Ratio of Si thickness to peak absorption wavelength</td>
<td>1/18</td>
<td>1/16</td>
<td>1/15</td>
<td>1/14.5</td>
</tr>
<tr>
<td>Skin depth in Al (nm)</td>
<td>12.0</td>
<td>11.6</td>
<td>12.0</td>
<td>13.2</td>
</tr>
<tr>
<td>Phase change at bottom Si-Al boundary (rad)</td>
<td>1.65</td>
<td>1.90</td>
<td>2.07</td>
<td>2.14</td>
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<td>Total phase change at two Si-Al boundaries (rad)</td>
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<td>2.47</td>
<td>2.75</td>
<td>2.91</td>
</tr>
</tbody>
</table>

To investigate angular dependence of light absorption, optical reflectivity spectra from fabricated devices were measured at different incident angles from 10° to 70° with 10° increment. Figures 2(a)-2(d) show the measurement results from devices of silicon cavity layer thickness of 30 nm, 40 nm, 50 nm, 60 nm respectively. It can be seen that large absorption greater than 90% is sustained up to 70° angle of incidence for all these devices. This explains why the colors of the devices do not change when they are viewed from large angles of incidence. This is because the round-trip phase delay inside the ultra-thin optical cavity is insensitive to the change of incident angle. The index of refraction of silicon is much higher than the index of refraction of air. The incident angle inside the silicon cavity is small according to the boundary condition (continuity of k-vector in the tangential direction). For incident angle of 70 degree in air, the incident angle in the silicon layer is about 17 degrees. The calculated round-trip phase delays at different incident angles are shown in Fig. 7 of the following section on simulation and discussion.
3. Simulation and discussion

In order to compare experimental results with simulation results, optical constants of Al and Si thin films were measured by using a variable angle spectroscopic ellipsometer (VASE by J. A. Woollam Company). The measured refractive index \( n \) and extinction coefficient \( k \) of Al and Si thin films are plotted in Figs. 3(a) and 3(b). Figure 3(a) shows that the 7 nm Al film has higher \( n \) and lower \( k \) compared to the 100 nm Al film. This is because the 7 nm Al film has small grains inside the film as shown in the inset of Fig. 1(b), and its thickness is above but close to the percolation threshold. The percolation threshold thickness is a critical thickness where the deposited metal film transforms its optical property from dielectric property to metallic property [28, 29]. Below the percolation threshold, the ultra-thin Al film consists of unconnected metal islands (clusters). Above the percolation threshold thickness, the metal islands (clusters) become connected and the film exhibits metallic property. By further increasing the film thickness, the film becomes a uniform film. Eventually, the optical property of sputtered film approaches to the optical property of bulk material. Therefore, the optical constants near the percolation threshold of Al film are strongly thickness-dependent as indicated in the measurement results shown in Fig. 3(a).

Fig. 2. (a-d) Measured optical reflectivity spectra from devices with Si cavity layer thicknesses from 30 nm to 60 nm at different angle of incidence form 10° to 70°.

Fig. 3. (a) Measured real and imaginary parts of the index of refraction of 7 nm and 100 nm aluminum films. The optical constants of Al films are dependent on the film thickness. (b) Measured real and imaginary parts of the index of refraction of 100 nm silicon film.
Using measured optical constants of Al and Si films shown in Figs. 3, optical reflectivity from the nanolayer structure is calculated by using the transfer matrix technique [30]. The optical reflectivity is

$$R = |r|^2 = \left| \frac{M_{21}}{M_{11}} \right|^2,$$

where $M_{21}$ and $M_{11}$ are elements of the transfer matrix,

$$M = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix} = D_0^{-1}(D_1 P D_1^{-1})(D_2 P D_2^{-1})D_3,$$  \hspace{1cm} (2)

In the transfer matrix above,

$$P_i = \begin{pmatrix} \exp(j\varphi) & 0 \\ 0 & \exp(-j\varphi_i) \end{pmatrix},$$  \hspace{1cm} (3)

where \(\varphi_i = \frac{2\pi(n_i - jk_i)d_i}{\lambda}\).

For TE polarization,

$$D_i = \begin{pmatrix} 1 & 0 \\ (n_i - jk_i)\cos\theta_i & (n_i - jk_i)\cos\theta_i \end{pmatrix},$$  \hspace{1cm} (4)

for TM polarization,

$$D_i = \begin{pmatrix} \cos\theta_i & \cos\theta_i \\ (n_i - jk_i) & -(n_i - jk_i) \end{pmatrix},$$  \hspace{1cm} (5)

where $i = 0, 1, 2, 3$ for air, top Al, Si, and bottom Al, respectively. In Eqs. (3)-(5), \(\theta_i\) is the incident angle in the medium $i$, and $n_i$ and $k_i$ are the real and imaginary parts of the refractive index of medium $i$, and \(\varphi_i\) is the propagation phase delay of optical wave in medium $i$. In Eq. (3), $d_1$ is the thickness of top Al film (7 nm), and $d_2$ is the thickness of Si cavity layer (30-60 nm).

Figure 4(a) shows the 2D plot of optical reflectivity vs. wavelength and Si cavity layer thickness, calculated by using the transfer matrix method with Eq. (1). Figure 4(b) shows the reflectivity spectra for silicon layer thicknesses of 30, 40, 50, and 60 nm with corresponding perfect absorption wavelengths of 570, 660, 750, and 840 nm, respectively. The calculated optical reflectivity spectra are in good agreement with the measured optical reflectivity spectra in Fig. 1(d).
To calculate the angular dependence of the absorption, the optical reflectivity spectra of four fabricated devices with 30, 40, 50, and 60 nm silicon layer thicknesses were calculated at different angles of incidence for TE and TM polarizations. For unpolarized incident light, reflectivity is taken as the average of the reflectivity of TE polarization and the reflectivity of TM polarization. Calculated reflectivity of unpolarized light versus wavelength and angle of incidence for structures with 30 nm to 60 nm silicon layer thicknesses are plotted and shown in Figs. 5(a)-5(d). It can be seen that perfect absorption wavelength does not change as angle of incidence increases from 0 to 70 degrees. The omni-directional absorption behavior is in good agreement with the measurement results.

![Fig. 5. Calculated optical reflectivity versus wavelength and angle of incidence for perfect light absorbers with Si cavity layer thicknesses of (a) 30 nm, (b) 40 nm, (c) 50 nm, and (d) 60 nm respectively. The absorption is insensitive to the change of incident angle up to 70°.](image)

To understand the perfect light absorption in the Al-Si-Al nanocavity, the round-trip phase delay in the nanocavity structure is calculated. For normal incidence, the electric field of the optical wave can be written as $E(z,t) = E_0 \exp(j(\omega t - 2\pi z/\lambda))$ in the air. Inside the silicon cavity, the propagation round trip phase delay is $\varphi_s = 4\pi n_2 d^2/\lambda$, where $n_2$ is the real part of the refractive index of silicon and $d_2$ is the thickness of silicon layer. The total round-trip phase delay, $\varphi_{\text{total}}$, is the sum of the propagation phase delay ($\varphi_s$) and phase delays caused by the reflection at the two cavity boundaries, i.e.

$$\varphi_{\text{total}} = \varphi_s - (\varphi_{21} + \varphi_{23})$$

where $\varphi_{21}$ and $\varphi_{23}$ are the phase changes due to reflection at the two boundaries with top Al layer and bottom Al, respectively. Figure 6(a) shows the propagation phase delays $\varphi_s$ in the silicon layer with thicknesses from 30 nm to 60 nm. Figure 6(b) shows the phase changes $\varphi_{21}$ and $\varphi_{23}$ at the two boundaries. $\varphi_{21}$ and $\varphi_{23}$ are the phase angles of the reflection coefficients $r_{21}$ and $r_{23}$ in the two silicon nanocavity boundaries. Figure 6(c) illustrates the total round trip phase delay $\varphi_{\text{total}}$ in the nanocavities of silicon layer thicknesses from 30 nm to 60 nm, calculated by using Eq. (6). In Fig. 6(c), it is shown that total round trip phase delay of zero occurs in the visible spectral range for silicon cavity thicknesses from 30 nm to 60 nm. The
zero round trip phase delay indicates that the zeroth order fundamental Fabry-Perot resonance mode causes perfect light absorption shown in Fig. 1(d).

To explain the angular insensitive behavior of the perfect light absorber, we calculated round-trip phase delay $\varphi_{\text{total}}$ inside the nanocavity at incident angle varying from 10° to 80° with 10° increment. Calculated phase delays vs. wavelength for different angles of incidence are shown in Figs. 7(a)-7(d) for nanocavities with different silicon layer thicknesses. It can be seen that the zero total round-trip phase delay is sustained at incident angles up to 70°. Therefore, the peak absorption wavelength corresponding to the zero total round-trip phase delay does not shift much across the spectrum as incident angle increases. This explains the angular insensitivity behavior of the perfect light absorbers.

To investigate where the optical energy is dissipated in the structure, we calculated electric and magnetic field distributions at perfect absorption wavelength of 750 nm by using
FDTD simulations (Lumerical Solution, Inc.). The electric field is plotted in the x-z plane and the magnetic field is plotted in the y-z plane. Figure 8(a) shows the real part of the x-component of the electric field in air and inside the device structure. In Fig. 8(a), the arrow indicates the direction of the electric field that is in the –x direction. It can be seen that the real part of the electric field holds the same direction as it propagates through the layer structure. Figure 8(b) shows the imaginary part of the electric field, and Fig. 8(c) shows the magnitude of the electric field. It can be seen in Fig. 8(c) that the electric field enhancement is small inside the structure and the electric field decays as it propagates in the structure. Figure 8(d) illustrates the real part of the y-component of the magnetic field profile. In Fig. 8(d), the arrows indicate the direction of the magnetic field. It is clearly seen that the magnetic field changes direction crossing the top Al-Si boundary. Figure 8(e) shows the imaginary part of the magnetic field, and Fig. 8(f) shows the amplitude of the magnetic field distribution in the structure. From Fig. 8(f), it can be seen that there is a strong magnetic field enhancement along the Si to bottom Al boundary. This strong magnetic field enhancement is caused by the anti-parallel electric dipole currents along the boundary of the aluminum metal and the silicon dielectric.

To investigate where exactly the optical energy is absorbed in the structure, we calculated energy absorption density $Q$ with the equation [31]

$$Q = \frac{\omega \epsilon_0}{8\pi} \text{Im} \left\{ \epsilon_r \right\} |E_o|^2,$$  \hspace{1cm} \text{(7)}

Fig. 8. Calculated real part, imaginary part, and magnitude of electric field (a-c) and magnetic field (d-f) distributions at the perfect absorption wavelength of 750 nm. The arrows indicate the directions of electric and magnetic fields in the structure.
where \( \text{Im}[\varepsilon_r] \) is the imaginary part of the electric permittivity of the medium, \(|E_0|\) is the amplitude of the electric field, and \( \omega \) is the angular frequency of the wave oscillation. Figure 9(a) shows calculated energy absorption density at 750 nm perfect absorption wavelength in the structure of 50 nm Si cavity layer bounded with 7 nm Al on top and 100 nm Al in bottom. The energy absorption density line profile in the z-direction is plotted in Fig. 9(b). In Fig. 9(b), the absorption energy, i.e. the area under the absorption curve, is 60% in the top Al layer, 5% in the Si layer, and 35% in the bottom Al metal. From Fig. 9(b), it is clearly seen that significant amount of energy is absorbed in the 7 nm Al layer on the top and the Al medium in the bottom. Also, it is seen that the maximal absorption density occurs in aluminum next to Si-aluminum boundaries. Only small portion of energy was absorbed in the silicon film.

![Image](image1)

**Fig. 9.** (a) Calculated energy absorption density at 750 nm perfect absorption wavelength in the device structure of 50 nm Si thickness bounded with 7 nm Al on the top and 100 nm Al in the bottom. (b) Absorption density profile at the perfect absorption wavelength of 750 nm.

To show potential biosensor application of the perfect light absorbers with percolation aluminum film, we applied liquid chemicals on top of a silicon device for measuring the change of its reflectivity spectrum. Figure 10 shows the measured reflectivity spectra from a device with 40 nm thick Si layer in air, and with applied IPA, methanol, and water on top. The measurement results show that by applying chemicals on the surface, the resonance wavelength has blue-shifts. The inset of Fig. 10 shows the photograph of the device partially covered by IPA on its surface. The color of the device changes from light blue to dark blue due to the presence of IPA liquid. The presence of IPA liquid reduces the reflectivity in the blue spectral range as shown in the Fig. 10.

![Image](image2)

**Fig. 10.** Measured optical reflectivity spectra from the 40 nm Si device in air, IPA, ethanol, and water. The inset shows the device picture when it is partially covered by IPA liquid and partially exposed to air.
4. Summary

In summary, nanocavity perfect light absorbers made of an ultra-thin percolation aluminum film and a high-index silicon thin film deposited on a thick aluminum film are demonstrated in visible and near-IR spectral range. The aluminum-silicon-aluminum thin film structure forms an asymmetric Fabry-Perot optical cavity. At the fundamental resonance mode where the total round trip phase delay is zero, incident optical wave is critically coupled into the nanocavity, resulting in complete light absorption in the structure. Using high-index silicon for the cavity medium enabled a significant reduction in cavity thickness to a factor of 18 with respect to the perfect absorption wavelength of 540 nm, about four times thinner than previously reported thin film cavity resonance enabled perfect light absorbers [16–19, 24]. Interestingly, it was found that the ratio of the cavity layer thickness to the perfect absorption wavelength decreases as the cavity layer thickness decreases due to increased phase delays in the silicon-aluminum boundaries at shorter wavelengths. Finally, optical energy absorption distribution in the nanostructure was calculated by simulations, and it was found that most energy was absorbed in the aluminum medium, not in the silicon cavity.

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