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## Thermodynamic limit to photonic-plasmonic light-trapping in thin films on metals

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We calculate the maximum optical absorptance enhancements in thin semiconductor films on metals due to structures that diffuse light and couple it to surface plasmon polaritons. The calculations can be used to estimate plasmonic effects on light-trapping in solar cells. The calculations are based on the statistical distribution of energy in the electromagnetic modes of the structure, which include surface plasmon polariton modes at the metal interface as well as the trapped waveguide modes in the film. The enhancement has the form  $4n^2 + n\lambda/h$  ( $n$  – film refractive index,  $\lambda$  – optical wavelength,  $h$  – film thickness), which is an increase beyond the non-plasmonic “classical” enhancement  $4n^2$ . Larger resonant enhancements occur for wavelengths near the surface plasmon frequency; these add up to 2 mA/cm<sup>2</sup> to the photocurrent of a solar cell based on a 500 nm film of crystalline silicon. We also calculated the effects of plasmon dissipation in the metal. Dissipation rates typical of silver reverse the resonant enhancement effect for silicon, but a non-resonant enhancement remains. © 2011 American Institute of Physics. [doi:10.1063/1.3658848]

### INTRODUCTION

The confinement of light by dielectric waveguides and by solar cells are companion fields that have been intensely studied for more than 30 years. The present paper is concerned with “light-trapping” in solar cells. Redfield was apparently the first to publish that guiding of scattered radiation by total internal reflection in a thin film would enhance absorption in solar cells.<sup>1</sup> Yablonoitch developed a calculation that quantified the maximal absorptance enhancement under the simplest, “classical” conditions as  $4n^2$ , where  $n$  is the index of refraction of the semiconductor film.<sup>2</sup> Yablonoitch assumed ergodicity, which is the requirement that all the electromagnetic modes within the film in a small frequency range have a common photon occupancy under steady illumination. For simplicity, he used the volume density-of-states of an infinite dielectric to obtain the  $4n^2$  result. The ergodic approach has been extended to accommodate more general forms for the photonic density of states, including exact treatments of waveguide modes,<sup>3</sup> and of multiple-layer dielectric structures.<sup>4,5</sup> Non-ergodic treatments of periodic gratings imposed on the metal and the thin-film layers have indicated light-trapping larger than inferred from the  $4n^2$  limit.<sup>6,7</sup>

In recent years there has been great interest in the possibility that electromagnetic excitations such as surface plasmons, localized or extended, might further facilitate light-trapping.<sup>8–11</sup> Zhou and Biswas reported calculations indicating enhancements beyond  $4n^2$  with a photonic crystal backreflector,<sup>12</sup> and recently Biswas and Xu reported that periodically patterned metal backreflectors also have enhancements beyond  $4n^2$ .<sup>13</sup> While photonic effects related to gratings, such as originally suggested by Sheng *et al.*,<sup>6</sup> might explain these enhancements, a second possibility is

that the sharing of electromagnetic energy between the surface plasmons and the waveguide modes of the thin film is involved.

In this paper we treat light-trapping in semiconductor films with metallic backreflectors, which is a simple model of a solar cell. We assume an ideal antireflection coating and a non-specific texturing of the interfaces. These assumptions are illustrated at the bottom of Fig. 1. We have extended the basic ergodic formalism to incorporate surface plasmon polaritons at the bottom, metal/semiconductor interface; the texturing is responsible for ergodic coupling of the photons and the surface plasmon polaritons (spp’s).

The essential concepts are illustrated at the top of Fig. 1. The sphere represents the possible wavevectors  $\vec{k}$  of photons with frequency  $\omega$  that are trapped within the film by total internal reflection; the film is assumed to be thick enough that effects of cutoff frequencies for the different waveguide modes can be neglected. The small missing portion at the top of the sphere corresponds to radiative modes that pass out of the film instead of being trapped. This part of the image is a graphical version of Yablonoitch’s original argument. Photons within the film are assumed to populate all portions of the spherical surface equally. Assuming that incident photons arrive through the air, the ratio of the surface area for the trapped wavevectors to the surface area for the radiative modes is  $4n^2$ —which is the “classical” enhancement effect.

The propagation wavevectors  $\vec{\beta}$  of the surface plasmon polariton (spp) modes at the same optical frequency  $\omega$  as the waveguide modes are represented by the planar “hoop” below the sphere; the area of this hoop indicates the areal density-of-states that is accessible to an electromagnetic quantum. It is evident that the area corresponding to trapped modes—waveguide and plasmonic—has increased relative to the area for radiative modes, which corresponds to an increased enhancement. This is the essential argument of the present paper.

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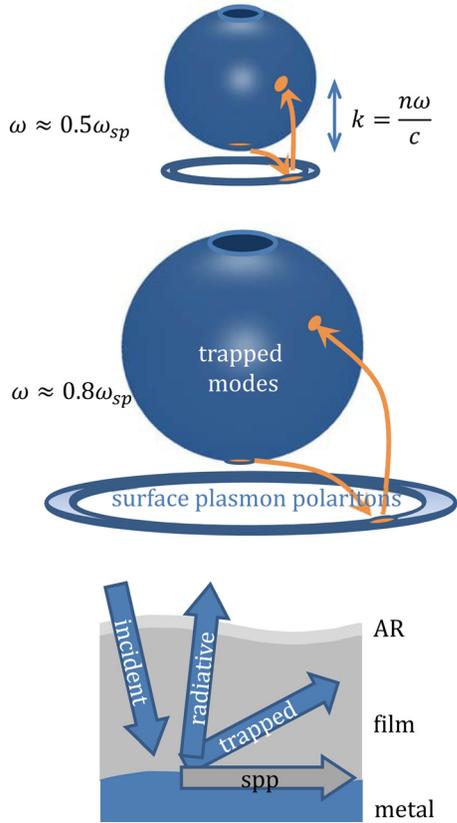


FIG. 1. (Color online) Ball and hoop drawings illustrating the electromagnetic modes for a thin dielectric film with a metallic backreflector. The spheres indicate photons with wavevector  $k$  lying in various directions. The dark “caps” represent radiative modes that are not trapped in the film; the rest of the sphere represents trapped photons. The hoops indicate the surface plasmon polaritons. Results are shown at two optical frequencies, one of which approaches the surface-plasmon resonance frequency  $\omega_{sp}$ . The surface areas of the elements are proportional to the trapped mode and surface plasmon polariton mode densities-of-states; light trapping may be considered as a random walk among these states, with escape when the quantum of energy reaches the radiative modes.

In the remainder of the paper, we implement these ideas for several cases. We first neglect plasmonic dissipation. We obtain relatively simple formulae for the absorptance of the semiconductor film in terms of general densities of trapped and spp modes and the “confinement factors” for the modes. For the case of surface plasmon polariton modes with frequencies well below the surface plasmon resonance, we find a simple “supraclassical” extension of the  $4n^2$  relation: The absorptance enhancement incorporating spp effects is  $4n^2 + n\lambda/h$ , where  $\lambda$  is the vacuum wavelength of the light, and  $h$  is the thickness of the film. We refer to this as the “non-resonant” case; remarkably, this limit is independent of the metal at the interface. The improvement beyond  $4n^2$  can be significant; for a 250 nm film, a 1000 nm wavelength, and  $n = 3.6$  (typical of silicon films), the spp’s increase the enhancement from 52 to 67. Since a 250 nm film is too thin to be well-approximated by the  $4n^2$  limit,<sup>3,7</sup> these numbers do need some refinement.

For photon frequencies that approach the surface plasmon resonance frequency, we obtain particular expressions using the Drude model to describe the dielectric properties of the metal. We present numerical calculations assuming

that the semiconductor is crystalline silicon. The resonant enhancements are larger than the non-resonant ones. For reference, we also calculate the short-circuit photocurrent density in corresponding solar cells under solar illumination, with the simplifying assumption that all photons absorbed in the silicon generate electron-hole pairs that are subsequently collected.

The absorptance spectra and the photocurrents may be compared with the recent electromagnetic simulations by Biswas and Xu that sought the optimum periodic structure of the metal/semiconductor interface for use in solar cells<sup>13</sup> and that achieved photocurrents higher than for the  $4n^2$  calculation. The magnitudes of their photocurrent calculations appear to be consistent with the calculations that we present, which (unsurprisingly) indicates that their enhancements beyond  $4n^2$  are partly plasmonic in origin.

In the final sections we incorporate plasmonic dissipation and give general results for absorptance in terms of photonic-plasmonic densities of states, confinement factors, and dissipation rates for the spp modes. We evaluate these for a Drude-Lorentz model of the metal and again illustrate the results numerically for thin silicon films on the metal. We find that a plasmon dissipation lifetime of thirty femtoseconds, typical of silver, reverses the resonant enhancement found with lossless surface plasmons. The non-resonant enhancement effect remains positive over a broader range of dissipations than does the resonant enhancement.

## THERMODYNAMIC ABSORPTANCE IN A THIN SEMICONDUCTOR FILM

In this paper we shall use the basic formalism developed by Stuart and Hall for their calculations of the thermodynamic limit for the absorptance of a thin semiconductor film<sup>3</sup>; they note that their approach is an application of the extended radiance theorem proven by Bassett.<sup>14</sup> In this section we briefly reprise some of their results that we use subsequently when surface plasmon polaritons are incorporated. Their equations (19) for the absorptance  $A$  of a semiconductor film are

$$A = \frac{\rho_{rad}}{\rho_{tot}} f_{rad} + \sum_m \frac{R_m}{h\rho_{tot}} f_m, \quad (1a)$$

$$\rho_{tot} = \rho_{rad} + \frac{1}{h} \sum_m R_m, \quad (1b)$$

where  $h$  is the thickness of the film.  $\rho_{rad}$  is the density of states (per unit volume and frequency) of the “radiative” photons within the film that are not trapped by total internal reflection.  $R_m$  is the density of states (per unit area and frequency) for a band  $m$  of trapped or waveguide modes within the device, and  $\rho_{tot}$  is the volume average of the density of electromagnetic modes in the film. In this model, the incident optical power is distributed evenly across all the electromagnetic modes of the film.  $f_{rad}$  and  $f_m$  are the fractions of the energy stored in a radiative or a waveguide mode that are absorbed by the semiconductor. Each mode stores the same energy, which is the ergodic assumption of this model.

Assuming that photons are incident on the device from air or vacuum with a unity index of refraction, Stuart and Hall's equation (18a) becomes

$$f_{\text{rad}} = \frac{\alpha h}{\alpha h + n\rho_0/(4\rho_{\text{tot}})}, \quad (1c)$$

where  $\alpha$  is the absorption coefficient of the film, and  $n$  is its index of refraction.  $\rho_0 \equiv \omega^2/\pi^2 c^3$  is the density-of-states for photons in air, including the two polarizations of light.

Stuart and Hall showed that these expressions yield what we shall term the classical value  $A_{cl}$  for the absorptance. For thicker films ( $h \gg \lambda/n$ ), the average volume density of states  $\rho_{\text{tot}}$  in the film including both the radiative and waveguide modes approaches the "classical" value:

$$\rho_{\text{tot}} \cong \rho_{cl} \equiv \frac{\omega^2 n^3}{\pi^2 c^3}. \quad (2a)$$

Additionally, the fraction  $f_m$  of the electromagnetic mode energy that is absorbed in the semiconductor approaches  $f_{\text{rad}}$  for nearly all modes; Eq. (1a) then yields for the total absorptance:

$$A_{cl} = \frac{4n^2 \alpha h}{1 + 4n^2 \alpha h}. \quad (2b)$$

$A_{cl}$  was originally proposed based on a less rigorous argument by Tiedje, Yablonovitch, Cody, and Brooks.<sup>15</sup> For  $\alpha h \ll 1$ , this expression reduces to  $A_{cl} = 4n^2 \alpha h$ , which is the "4n<sup>2</sup>" limit proposed by Yablonovitch<sup>2</sup>; we define the "classical" enhancement factor  $Y_{cl} = 4n^2$ .

### THERMODYNAMIC ABSORPTANCE INCORPORATING A LOSSLESS SURFACE PLASMON POLARITONS

Stuart and Hall's treatment can be adapted for a band of surface plasmon polaritons with areal density  $R_{spp}$ ; we neglect dissipation by the metal in this section and treat lossy spp's later. For strictly planar interfaces, spp's do not couple to the radiative and waveguide modes; one may think of two independent spaces of modes—one for the semiconductor waveguide, and one for the surface plasmon polaritons—that are uncoupled.<sup>16,17</sup> When the interface between the semiconductor and the metal becomes textured, the two types of mode become coupled. This coupling has been extensively studied; all that is needed for the present calculation is that the coupling be strong enough that electromagnetic energy is shared evenly between the radiative, waveguide, and spp modes at an optical frequency  $\omega$ ; we will return to this subject briefly when we discuss lifetime broadening for the spp modes.

We denote the areal energy density that is stored in a specific electromagnetic mode  $m$  as  $u_m$ . The semiconductor film absorbs power from this mode at the rate  $r_m = \gamma \Gamma_m u_m$ .  $\Gamma_m$  is the "confinement factor" for a mode, which is the fraction of the electromagnetic energy in the mode that lies within the semiconductor film; the remainder lies within the metal at the back of the film or in a dielectric or in air at the top of the film. The value of  $\gamma$  is related to the absorption coefficient of the semiconductor by the equation

$$\gamma \equiv \alpha c/n, \quad (3)$$

which is obtained by consideration of the radiative modes. The fractional absorption  $f_m$  due to the trapped modes of the film—waveguide or spp—then has the form

$$f_m = \frac{\Gamma_m \gamma}{\Gamma_m \gamma + \rho_0 c/(4h\rho_{\text{tot}})} = \frac{\Gamma_m \alpha h}{\Gamma_m \alpha h + n\rho_0/(4\rho_{\text{tot}})}. \quad (4)$$

This expression is based on Eq. (18b) of Stuart and Hall's paper,<sup>3</sup> but is simpler because of a somewhat different interpretation of absorption coefficients  $\alpha$ . This issue is treated in the Appendix. The form for  $f_{\text{rad}}$  (Eq. (1c)) is unchanged.

For completeness, we write the corresponding version of Eq. (1):

$$A_{spp} = \frac{\rho_{\text{rad}}}{\rho_{\text{tot}}} f_{\text{rad}} + \sum_m \frac{R_m}{h\rho_{\text{tot}}} f_m + \frac{R_{spp}}{h\rho_{\text{tot}}} f_{spp}. \quad (5a)$$

Equations (4) and (5) are a complete description as long as we can neglect losses to the metal backreflector or direct decay of the waveguide modes by radiation; we generalize Eq. (5) in a subsequent section. Although it will not be needed in the present paper, where we use the classical approximation for the waveguide modes, Stuart and Hall's definition of  $\rho_{\text{rad}}$  (their Eq. (2)) reduces to the following form when the top interface is with air<sup>3</sup>

$$\rho_{\text{rad}} = \left(1 - \sqrt{1 - 1/n^2}\right) \rho_{cl}. \quad (5b)$$

### Non-resonant enhancement by surface plasmon polaritons

One complication in using Eq. (5) is the presence of the confinement factors in Eq. (4). For the classical 4n<sup>2</sup> enhancement, the confinement factors  $\Gamma_m$  for the waveguide modes are unity, which requires a sample of thickness  $h \gg \lambda/2n$ ; this yields  $f_m = f_{\text{rad}}$ , and what we will call the supraclassical absorptance

$$A_{scl} = f_{\text{rad}} \frac{\rho_{cl}}{\rho_{\text{tot}}} + f_{spp} \frac{R_{spp}}{h\rho_{\text{tot}}}. \quad (6)$$

A similar simplification occurs for the surface plasmon polariton modes when the optical frequency for an experiment lies well below the surface plasmon resonance frequency. In this case the spp confinement factor also approaches unity (i.e.,  $f_{spp} = f_{\text{rad}}$ ), as we will show shortly. With these simplifications, we can factor  $f_{\text{rad}}$  out of the absorptance expression (5). The total absorptance is

$$A_{nr} = f_{\text{rad}} \left( \frac{\rho_{\text{rad}}}{\rho_{\text{tot}}} + \sum_m \frac{R_m}{h\rho_{\text{tot}}} + \frac{R_{spp}}{h\rho_{\text{tot}}} \right) = f_{\text{rad}} (\Gamma_m, \Gamma_{spp} = 1). \quad (7)$$

For the classical limit we are using the thick-sample approximation for the total density of photonic states (the sum of the radiative and the dielectric waveguide modes)

$$\rho_{rad} + \sum_m \frac{R_m}{h} \cong \rho_{cl} \equiv \frac{\omega^2 n^3}{\pi^2 c^3}.$$

The “supraclassical” density is then  $\rho_{scl} \equiv \rho_{cl} + R_{spp}/h$ . We obtain the following expression for the non-resonant absorptance  $A_{nr} = f_{rad}$  by evaluating Eq. (1c):

$$A_{nr} = \frac{4n^2 \alpha h (1 + R_{spp}/(h\rho_{cl}))}{1 + 4n^2 \alpha h (1 + R_{spp}/(h\rho_{cl}))}. \quad (8a)$$

The classical enhancement  $Y_{cl} = 4n^2$  is now increased to an spp-enhanced value:

$$Y_{nr} = 4n^2 (1 + R_{spp}/(h\rho_{cl})). \quad (8b)$$

In the non-resonant regime, spp’s have essentially the same dispersion relation as the photons in the thin film; they are like a two-dimensional photon gas confined to the metal/semiconductor interface and moving at a speed of  $c/n$ .<sup>16</sup> The non-resonant density of spp states is then

$$R_{spp}^{nr} = \frac{n^2 \omega}{2\pi c^2}. \quad (9a)$$

Since  $\rho_{cl} = \omega^2 n^3 / \pi^2 c^3$ , we obtain the non-resonant enhancement factor  $Y_{nr}$  incorporating spp’s by substitution into Eq. (8b)

$$Y_{nr} = 4n^2 + (n\lambda/h). \quad (9b)$$

Remarkably, this expression does not depend on the metal’s properties. Figure 2 shows calculations for three films of c-Si based on the latter’s measured refractive index.<sup>18</sup> The spp’s increase the enhancement substantially for thinner films, but for these the  $4n^2$  approximation is not accurate. A similar effect would obtain in a-Si:H, which has essentially the same refractive index at this wavelength. The relative gain over  $4n^2$  increases in low-index materials such as organic semiconductors.<sup>4,10</sup>

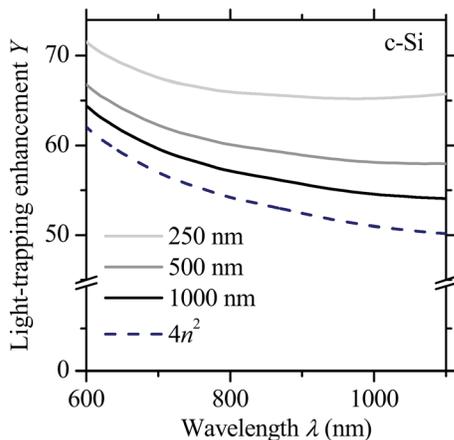


FIG. 2. (Color online) Light-trapping enhancement factor  $Y$  as a function of wavelength for light absorption by thin films of silicon with a metal back-reflector; three thicknesses are shown. The curve labeled  $4n^2$  is the classical enhancement due to waveguide trapping. It does not include surface plasmon polaritons (spp’s), which cause a non-resonant enhancement beyond  $4n^2$ .

## Surface plasmon polariton density at a planar interface

The last section evaluated  $R_{spp}$  in the low-frequency regime; more generally it must be evaluated using the full dispersion relation for the in-plane propagation wavevector  $\beta(\omega)$  of spp’s (Ref. 16)

$$\beta^2 = k_0^2 \frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2}, \quad (10)$$

where  $k_0 = \omega/c$  is the vacuum wavenumber for an electromagnetic wave,  $\epsilon_1(\omega)$  is the relative dielectric function of the metal, and  $\epsilon_2(\omega)$  is the dielectric function of the semiconductor.  $\epsilon_1$  and  $\epsilon_2$  are often complex; when both are real,  $\beta^2$  diverges at the surface plasmon frequency  $\omega_{sp}$  defined implicitly by  $\epsilon_1(\omega_{sp}) + \epsilon_2(\omega_{sp}) = 0$ .

With typical dielectric functions at lower frequencies  $\omega$ ,  $\beta^2$  is nearly real, positive, and monotonically increasing.  $R_{spp}$  is then evaluated using a textbook procedure. The total density  $N_{spp}(\omega)$  of spp states (per unit area of interface) having frequencies less than some value  $\omega$  is given by the expression

$$N_{spp}(\omega) = \left(\frac{1}{2\pi}\right)^2 (\pi\beta^2), \quad (11)$$

where  $(1/2\pi)^2$  is the density of possible eigenvectors  $\beta$  in the two-dimensional space of spp wavevectors. The density of surface plasmon polaritons  $R_{spp}(\omega)$  (per unit area and frequency) is obtained from

$$R_{spp}(\omega) = \frac{\partial N_{spp}}{\partial \omega} = \frac{1}{4\pi} \frac{d}{d\omega} (\beta^2). \quad (12)$$

## Drude model for surface plasmon polaritons

We analyze the spp contribution to the absorptance in more detail using the Drude-Lorentz model for  $\epsilon_1$  (Ref. 16)

$$\epsilon_1 = \epsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\gamma_p \omega}, \quad (13)$$

where  $i = \sqrt{-1}$ ,  $\omega_p$  is the bulk plasmon frequency, and  $\gamma_p$  is the plasmon thermalization frequency. In this section we assume that  $\gamma_p = 0$ . The density of surface plasmon polaritons  $R_{spp}(\omega)$  (per unit area and frequency) is then

$$R_{spp}(\omega) = \frac{\partial N_{spp}}{\partial \omega} = \frac{\beta^2}{2\pi\omega} \left(1 + \frac{\omega_p^2 \epsilon_2}{\omega^2 \epsilon_1 (\epsilon_1 + \epsilon_2)}\right), \quad (14)$$

where we neglect a term corresponding to the dispersion of the semiconductor  $d\epsilon_2/d\omega$ .

In addition to the density-of-states we will need the confinement factor  $\Gamma_{spp}$  of an spp, which is the fraction of its electromagnetic energy that lies in the dielectric or semiconductor; this is related to the wavevectors  $k_1$  and  $k_2$  describing the evanescent decay of the electromagnetic field amplitude

into the metal and the dielectric, respectively, which we take over from Maier's textbook (equations (2.10)-(2.14))<sup>17</sup>

$$k_1^2 = \beta^2 - \varepsilon_1(\omega^2/c^2),$$

$$k_2^2 = \beta^2 - \varepsilon_2(\omega^2/c^2).$$

Squaring each component of the electric field and summing obtains the amplitude<sup>17</sup>

$$E_1^2 = C^2 \frac{k_1^2 + \beta^2}{\varepsilon_1^2} \exp(2k_1 z) \quad (z < 0),$$

$$E_2^2 = C^2 \frac{k_2^2 + \beta^2}{\varepsilon_2^2} \exp(-2k_2 z) \quad (z > 0),$$

where  $C$  is an arbitrary amplitude. Using the expression for the time-averaged electromagnetic energy density  $u = (\varepsilon_0/16\pi) E^2(d/d\omega)(\omega\varepsilon)$ ,<sup>19</sup> and integrating out from the interface to obtain an areal energy density, we obtain

$$U_1 = \frac{\varepsilon_0}{16\pi} \left( \varepsilon_\infty + \frac{\omega_p^2}{\omega^2} \right) C^2 \frac{k_1^2 + \beta^2}{2\varepsilon_1^2 k_1}, \quad (15a)$$

$$U_2 = \frac{\varepsilon_0}{16\pi} C^2 \frac{k_2^2 + \beta^2}{2\varepsilon_2 k_2}, \quad (15b)$$

where we have again neglected the dispersion of the semiconductor  $d\varepsilon_2/d\omega$ . The confinement factor  $\Gamma_{spp}$  is defined as the ratio

$$\Gamma_{spp} = \frac{U_2}{U_1 + U_2}. \quad (15c)$$

In Fig. 3 we graph  $R_{spp}$  using parameters for silver:  $\omega_p = 1.3 \times 10^{16} \text{ s}^{-1}$ ,  $\varepsilon_\infty = 3.3$ .<sup>20-22</sup> We used the measured dielectric function for c-Si,<sup>18</sup> which is complex and prevents divergence of the expression for  $R_{spp}$  near the surface plasmon resonance frequency of  $\omega_{sp} = 3.0 \times 10^{15} \text{ s}^{-1}$ . We set  $R_{spp} = 0$  for  $\text{Re}(\beta^2) < 0$ .

We convolved  $R_{spp}$  with a Lorentzian representing a  $\tau_R = 20$  fs lifetime broadening to account for radiation of the spp energy back into waveguide or radiative modes. The present formalism that assumes ergodic contact between the waveguide modes, spp's, and radiative modes requires that this lifetime be much shorter than the residence time of a photon in the cell. The residence time is roughly  $4n^2(hn/c)$ , which is about 0.6 ps in a silicon cell of one micron thickness. Experimental measurements of the radiative lifetime for spherical silver nanoparticles in fluid ( $n = 1.5$ ) are in the range 1–6 fs for particle sizes 150 nm–20 nm.<sup>23</sup>

### Resonant enhancement of absorptance of a silicon film

In Fig. 4, the lower panel shows the numerical calculation for the absorptance of a 500 nm film based on the supra-classical Eq. (6) above for a series of metal interfaces with differing surface plasmon frequencies and using the measured optical properties of c-Si.<sup>18</sup> The three different curves

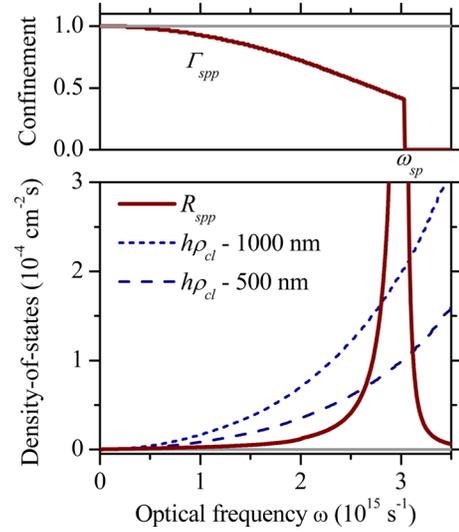


FIG. 3. (Color online) (Lower) Areal state density  $R_{spp}$  as a function of optical frequency for surface plasmon polaritons (spp's) at the silver/silicon interface; the surface plasmon frequency  $\omega_{sp}$  is marked on the top axis. The areal densities of trapped waveguide modes for 500 nm and 1000 nm thick silicon films are also illustrated. (Upper) The spp confinement factor  $\Gamma_{spp}$  as a function of frequency;  $\Gamma_{spp}$  is the fraction of the energy in an spp that lies in the semiconductor. Confinement factors  $\Gamma_m$  are assumed to be unity for the waveguide modes.

were calculated by varying the bulk plasmon frequency parameter  $\omega_p$  of the metal to obtain different surface plasmon frequencies  $\omega_{sp}$ . Each of the absorptance curves shows a feature that is close to the wavelength of its surface plasmon. The classical absorptance  $A_{cl}$  (Eq. (2b)) is also graphed and forms the lower envelope of the curves. The 20 fs lifetime broadening noted earlier causes a small absorptance at wavelengths longer than the bandgap threshold for c-Si, which is at about 1100 nm.

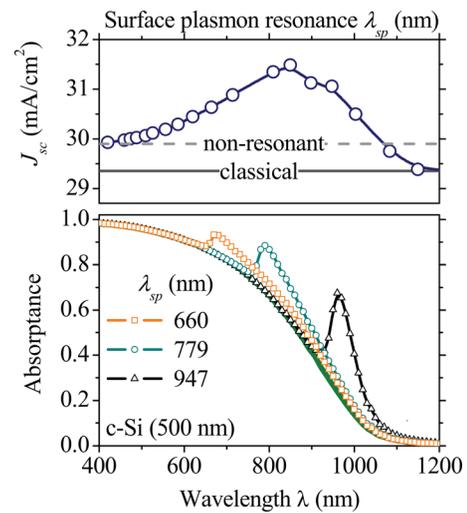


FIG. 4. (Color online) (Lower panel) Supra-classical absorptance spectra calculated for 500 nm films of c-Si incorporating lossless surface plasmon polaritons with three different surface plasmon frequencies; the classical,  $4n^2$  absorptance is also drawn and is the lower envelope of the supra-classical curves. (Upper) Short-circuit photocurrent densities  $J_{sc}$  under solar illumination for a solar cell based on a 500 nm silicon film; the values are calculated using absorptance curves for a wide range of surface plasmon resonance wavelengths.

The upper panel shows the short-circuit current densities  $J_{SC}$  calculated for the corresponding crystalline silicon solar cells.  $J_{SC}$  is calculated by integrating the product of the absorptance, the photon flux spectrum derived from a standard for solar illumination<sup>24</sup> and the electronic charge. In physical solar cells current densities are lower than the value calculated from these absorptances because of imperfect antireflection coatings, absorption by the doped layers that does not contribute to the photocurrent, photocarrier recombination, etc.. The horizontal lines are independent calculations based on absorptances for classical “ $Y_{cl} = 4n^2$ ” enhancement (Eq. (2)) and for the non-resonant spp enhancement  $Y_{nr} = 4n^2 + n\lambda/h$  (Eq. (9b)).

For the shortest surface plasmon wavelengths,  $J_{SC}$  approaches the non-resonant enhanced value of 30 mA/cm<sup>2</sup>. The nearly 20% increase in the enhancement factor (cf. Fig. 2), beyond its classical value of  $4n^2$ , integrates out to just a 2% increase in  $J_{SC}$ . As the surface plasmon resonance shifts to the center of the graph, the resonant enhancement increases the current to a maximum value near 31.5 mA/cm<sup>2</sup>. The enhancement maximum occurs near  $\lambda_{sp} \sim 850$  nm because that is the wavelength for which the classical absorptance for a 500 nm film is about 0.5, as can be verified by examining the figure. Enhancements at shorter wavelengths do not much affect the photocurrent, since all those photons are absorbed even without enhancement. At longer wavelengths, little photocurrent is collected with or without enhancement. As the spp band shifts to still longer wavelengths, the photocurrent falls to 29.3 mA/cm<sup>2</sup>, which is the value for classical enhancement  $4n^2$ .

Biswas and Xu have recently published full electromagnetic calculations of absorptances for silicon on silver structures that can be compared with these results.<sup>13</sup> Their calculations assumed a particular type of profile for periodic structuring of this interface, and they calculated the absorptances for a wide range of profile parameters to identify an optimum shaping. They also assumed negligible plasmonic loss. For a 500 nm c-Si film, they report 3 mA/cm<sup>2</sup> enhancement beyond the classical calculation that is due to optimized interface structure. The present calculation yielded about 2 mA/cm<sup>2</sup> of increase (cf. Fig. 4). We speculate that the specific texture they applied moved the surface plasmon resonance from its planar value of 630 nm (cf. Fig. 3) out to about 850 nm; this is a well-established effect.<sup>16</sup> We think this effect would yield about the same 2 mA/cm<sup>2</sup> that we found by adjusting the Drude parameters of the metal interface for the ergodic model, so the plasmonic effect is responsible for 2/3 of their reported enhancement. The remaining 1 mA/cm<sup>2</sup> of enhancement they report is presumably due to the additional grating-effects proposed by Sheng *et al.*<sup>6</sup> and Yu *et al.*<sup>7</sup>

Yu *et al.* and also Green have recently published proposals to use evanescent modes at the interface between two dielectrics to exceed the  $4n^2$  limit.<sup>4,7</sup> A high-index, transparent material is placed in contact with a low-index absorbing material, and photons trapped in the high-index material couple to the absorber layer via the evanescent wave extending into it. This proposal is related conceptually to the present calculations, which involve an evanescent wave extending

into the c-Si from a metal interface, and it avoids the plasmonic dissipation that we explore in the next section.

## THERMODYNAMIC ABSORPTANCE INCORPORATING LOSSY SURFACE PLASMON POLARITONS

The previous section assumed that the non-radiative electromagnetic modes of the semiconductor/metal structure lose energy only by semiconductor absorption; of course the structure as a whole also leaks energy because of the coupling to its radiative modes. In this section we introduce intrinsic loss by an spp through the plasmon damping of the metal (the  $\gamma_p$  term of the Drude-Lorentz model Eq. (10)). The same formalism could be used to treat leaking of the waveguide modes through the top interface of the semiconductor film, which is present in principle due to semiconductor absorption and is increased by texturing or corrugation of the top interface.<sup>25</sup> Similarly, for a planar backreflector interface, the finite conductivity of a metal at optical frequencies leads to attenuation of the waveguide modes<sup>25</sup>; our estimate of this latter effect is that it is about a hundred times slower than the loss of energy to the radiative modes, which renders it negligible.

We write for the total fractional absorption of the surface plasmon polaritons

$$f'_{spp} = \frac{\Gamma_{spp}\gamma + \gamma_{spp}}{\Gamma_{spp}\gamma + \gamma_{spp} + \frac{\rho_0 c}{4h\rho_{tot}}}, \quad (16)$$

where  $\gamma = \alpha n/c$  (as before), and  $\gamma_{spp}$  and  $\Gamma_{spp}$  are the dissipation rate and the confinement factor for the spp's. For convenience we define the leakage rate for the emission of electromagnetic energy through the radiative modes

$$\gamma_{rad} = \frac{\rho_0 c}{4h\rho_{tot}},$$

The total absorptance of the system is then

$$A_{tot} = \frac{\rho_{rad}}{\rho_{tot}} f_{rad} + \sum_m \frac{R_m}{h\rho_{tot}} f_m + \frac{R_{spp}}{h\rho_{tot}} f'_{spp}, \quad (17)$$

where  $f_{rad}$  and  $f_m$  retain the previous definitions (Eq. (1c) and Eq. (4)). In Eq. (16), only the term  $\Gamma_{spp}\gamma$  corresponds to absorption in the semiconductor; we write the partial absorptance by the semiconductor film as

$$A'_{spp} = \frac{\rho_{rad}}{\rho_{tot}} f_{rad} + \sum_m \frac{R_m}{h\rho_{tot}} f_m + \frac{R_{spp}}{h\rho_{tot}} \frac{\Gamma_{spp}\gamma}{\Gamma_{spp}\gamma + \gamma_{spp} + \gamma_{rad}}, \quad (18)$$

which is the lossy version of Eq. (5) above. Assuming the waveguide modes can be treated in the classical limit ( $\Gamma_m = 1$ , etc.), we obtain the general “supraclassical” expression including plasmonic losses that is analogous to Eq. (6) above

$$A'_{scl} = f_{rad} \frac{\rho_{cl}}{\rho_{tot}} + \frac{R_{spp}}{h\rho_{tot}} \frac{\Gamma_{spp}\gamma}{\Gamma_{spp}\gamma + \gamma_{spp} + \gamma_{rad}}. \quad (19)$$

### Drude-Lorentz model for the lossy surface plasmon polariton

The density-of-states  $R_{spp}$  in Fig. 3 is not changed markedly by the incorporation of a non-zero value for  $\gamma_p$ . Within the Drude-Lorentz model, using Eq. (19) above for  $A'_{scl}$  requires that we establish the relationship between  $\gamma_{spp}$  and the Drude-Lorentz parameter  $\gamma_p$ . The procedure we use is related to the discussion in the Appendix, where we calculate the dissipation rate for a waveguide mode from the product of its spatial attenuation constant and the mode's group velocity.

For spp's, we first calculated the attenuation constant  $\alpha_{spp}$  using the complex propagation wavevector  $\beta$

$$\alpha_{spp} = 2\text{Im}(\beta). \quad (20a)$$

Along the interface, the field amplitude decays exponentially with attenuation coefficient  $\text{Im}(\beta)$ ; this is doubled to obtain the power attenuation coefficient, which is proportional to the field amplitude squared. The generalization of Eq. (3) ( $\gamma = \alpha c/n$ ) to propagating surface plasmon polaritons is based on evaluating the group velocity  $v_g^{spp}$  of an spp

$$\gamma_{spp} = v_g^{spp} \alpha_{spp}, \quad (20b)$$

$$v_g^{spp} \equiv \left( \frac{d\beta}{d\omega} \right)^{-1}.$$

If the spp density-of-states  $R_{spp}$  has been evaluated, one can use the relation (cf. Eq. (12))

$$v_g^{spp} = \frac{2\beta}{d(\beta^2)} = \frac{\beta}{2\pi R_{spp}}. \quad (20c)$$

We have previously evaluated  $R_{spp}$  from the Drude-Lorentz form for  $\epsilon_1$  to obtain Eq. (14), and of course Eq. (10) still obtains for  $\beta$ .

This procedure has a difficulty; Eq. (20b) assumes that the dissipation rate of the spp in the semiconductor layer ( $\gamma$ ) is negligible, which is not true in the limit of small  $\gamma_p$ . We therefore evaluated  $\gamma_{spp}$  for "lossless" silicon ( $\gamma = 0$ ); the procedure assumes that dissipation does not significantly alter the spp's properties (cf.  $\beta(\omega)$ ), which we found to be reasonably correct for  $\gamma_p$  up to  $3 \times 10^{13} \text{ s}^{-1}$  (close to measurements for silver). In Fig. 5 we have illustrated the calculations for  $\gamma_{spp}$  as a function of optical frequency  $\omega$ , along with the measurements of  $\gamma$  for c-Si.  $\gamma_{spp}$  rises essentially as  $\omega^2$ , reaching  $\gamma_p$  at  $\omega = \omega_{sp}$ .  $\gamma$  falls much faster, since it has a threshold near the bandgap for c-Si.

Using Eqs. (15) for the supraclassical absorptance  $A'_{scl}$  and Eq. (20) for  $\gamma_{spp}$ , we did a numerical evaluation of the effects of dissipation on the non-resonant enhancement factor  $Y_{nr}$ . We chose conditions of technical interest for c-Si solar cells:  $\lambda_{sp} = 530 \text{ nm}$ ,  $\lambda = 1000 \text{ nm}$ ; the other details are the same as for the absorptance spectra in Fig. 4. The enhancement factor was estimated from the ratio  $A'_{scl}/\alpha h$  at  $\lambda = 1000 \text{ nm}$ .

We present these calculations as Fig. 6 for two film thicknesses. For the 500 nm film the enhancement due to surface

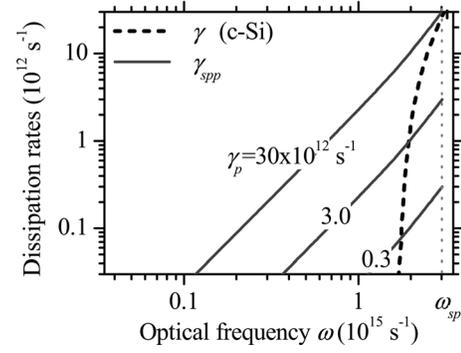


FIG. 5. Dissipation rates for electromagnetic modes as a function of optical frequency. The solid lines show surface plasmon polariton dissipation rates  $\gamma_{spp}$  based on the Drude Lorentz model ( $\omega_p = 1.3 \times 10^{16} \text{ s}^{-1}$ ) for the metal and the measured dielectric function of c-Si; the corresponding surface plasmon frequency  $\omega_{sp} = 3.0 \times 10^{15} \text{ s}^{-1}$  is marked. Results are shown for three plasmon dissipation rate parameters  $\gamma_p$ . The dashed line corresponds to inter-band absorption in c-Si and is based on absorption coefficient measurements.

plasmons is essentially canceled when  $\gamma_p = 3 \times 10^{13} \text{ s}^{-1}$ , which is close to experimental estimates of this parameter in silver.<sup>20,21</sup> For the 250 nm film the enhancement is more robust, which could be of technological interest. For a sample this thin classical expression ( $4n^2$ ) needs some adjustment for waveguide mode cutoff effects.<sup>3,7</sup>

In Fig. 7 we present our calculations for the full absorptance spectra  $A'_{scl}$  for a series of values of  $\gamma_p$ ; we chose the plasma frequency to obtain a surface plasmon resonance at  $\lambda_{sp} = 853 \text{ nm}$ , corresponding to the enhancement maximum in Fig. 4. For plasmon damping rates greater than  $\gamma_p = 1 \times 10^{12} \text{ s}^{-1}$ , plasmonic losses are greater than the gain. The non-resonant enhancements graphed in Fig. 5 remained positive through about  $\gamma_p = 3 \times 10^{13} \text{ s}^{-1}$ , which suggests that the non-resonant effect is much less influenced by plasmon dissipation than is the resonant effect.

These results are not unexpected. For the non-resonant case, most of an spp's energy lies in the semiconductor and is not subject to plasmon dissipation. However, resonant enhancements occur when the surface plasmon polariton has

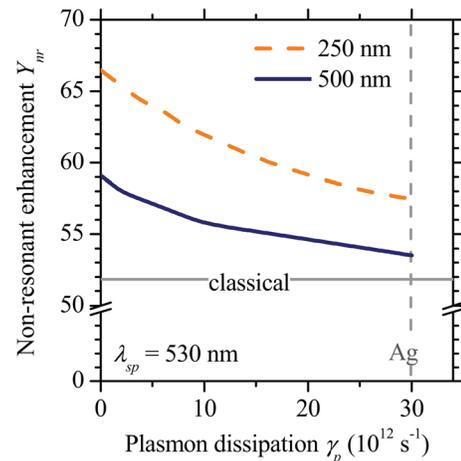


FIG. 6. (Color online) Effect of the plasmon dissipation rate  $\gamma_p$  on the non-resonant light-trapping enhancement factor  $Y_{nr}$  for silicon films of two thicknesses and evaluated at a wavelength of 1000 nm; the underlying surface plasmon resonance wavelength, which does affect the dissipation, is also shown. The classical enhancement  $Y = 4n^2$  is also shown.

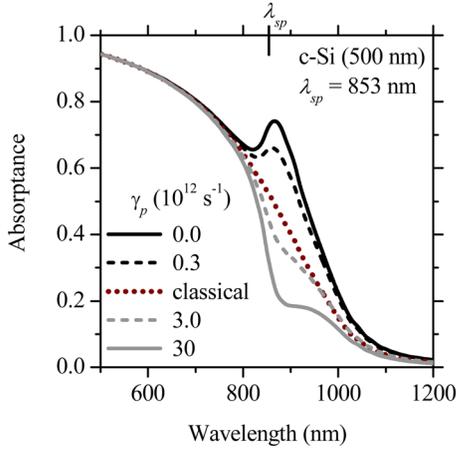


FIG. 7. (Color online) Calculated partial absorbance spectra of a 500 nm silicon film on a backreflector with surface plasmon resonance at 853 nm. Results are shown for four values of the Drude-Lorentz plasmon dissipation rate  $\gamma_p$ . The classical absorbance spectrum without surface plasmons is also illustrated.

about half its energy within the metal (cf. Fig. 3), and it is this fraction of the energy that is subject to plasmonic loss. In order to realize significant enhancements in the resonant regime, this dissipation must be slower than the fundamental semiconductor absorption ( $\gamma_p \ll \gamma$ ). In c-Si, the interband absorption rate at a wavelength of 900 nm can be evaluated from the absorption coefficient to obtain  $\gamma = 3 \times 10^{12} \text{ s}^{-1}$ ; it is thus unsurprising that plasmonic loss rates comparable to this will suppress the resonant enhancement effect of increased semiconductor absorption.

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## APPENDIX A: DIELECTRIC ABSORPTION FOR WAVEGUIDE MODES AND SPP'S

A central issue in applying optical waveguide calculations to solar cells is evaluating the power absorbed by the semiconductor from a density of electromagnetic energy  $u_m$  (per unit area of film) that is stored in a given mode  $m$ . Ordinary optical spectroscopy measures the absorption coefficient  $\alpha(\omega)$ , which is the attenuation coefficient for a radiative mode traveling at group velocity  $v_g^0 = c/n$ . The volume rate of absorption of electromagnetic energy for radiative modes is then  $P_{abs} = \alpha v_g^0 u_{rad} = (\alpha c/n) u_{rad}$ , where  $u_{rad}$  is the energy stored in the radiative modes of the film.

We write the power density  $r_m$  that is absorbed by the semiconductor film from the energy density  $u_m$  in a waveguide mode as

$$r_m = \Gamma_m \alpha v_g^0 u_m, \quad (\text{A1})$$

where  $\alpha$  is the bulk absorption coefficient of the semiconductor, and  $\Gamma_m$  is the energy confinement factor for the mode. This equation is a statement that the rate of energy absorption by the semiconductor is proportional to the electromagnetic energy density, and that the proportionality constant is the same for guided modes and radiative modes.

A version of Eq. (A1) was used in the recent paper by Yu, Raman, and Fan.<sup>7</sup> However, Stuart and Hall wrote  $r_m = \Gamma_m \alpha v_g^m u_m$  (their Equation (15));  $v_g^m$  is the group velocity of mode  $m$ , not the group velocity for a radiative mode.<sup>3</sup> As a consequence, their expression (18b) for  $f_m$  (the fractional absorption) contains  $v_g^m/v_g^0$ , which is the ratio of the group velocity of the waveguide mode to the ordinary speed of light in a material of index  $n$ . With our interpretation, the ratio is  $v_g^0/v_g^0$ , which of course simplifies their expression to the one we've used (Eq. (4) above).

Textbook summaries of loss calculations in dielectric waveguides are not clear on this point.<sup>25,26</sup> Group velocities change rapidly near waveguide cutoff frequencies, but for dielectric waveguides there is also a rapid loss of confinement in this frequency region that complicates the interpretation of attenuation.

We thus present an elementary waveguide calculation for a plane parallel, ideal metal wall waveguide. This avoids the issue of confinement factors and seems to us sufficient to confirm that Eq. (A1) is correct. This calculation is an extension of a textbook example<sup>25</sup> and is also the basis for our method for evaluating the spp dissipation rate  $\gamma_{spp}$ .

The transverse electric (or magnetic) modes of the waveguide have the dispersion relation

$$\begin{aligned} \beta_N^2 &= k^2(n + iK)^2 - ((N + 1)\pi/h)^2 \\ &\cong k^2 n^2 - ((N + 1)\pi/h)^2 + 2ik^2 nK, \end{aligned} \quad (\text{A2})$$

where the mode index  $N$  is a positive integer, the complex index of refraction of the dielectric is  $n + iK$ , the mirror separation is  $h$ , and  $k = \omega/c$ . We have assumed that  $K$  is small and keep only terms to first order in  $K$ .

The real part of  $\beta_N$  is the propagation constant for the waveguide mode, and the imaginary part is the exponential decay constant for the field in the guided mode. We obtain

$$\text{Re}(\beta_N) \cong \sqrt{k^2 n^2 - ((N + 1)\pi/h)^2}, \quad (\text{A3})$$

$$\text{Im}(\beta_N) \cong k^2 nK / \text{Re}(\beta_N). \quad (\text{A4})$$

The intensity attenuation constant  $\alpha_N$  in this mode is

$$\alpha_N = 2\text{Im}(\beta_N) = 2k^2 nK / \text{Re}(\beta_N). \quad (\text{A5})$$

Using the result  $\alpha = 2Kk$  connecting the ordinary optical absorption coefficient  $\alpha$  and  $K$  for the dielectric,<sup>25</sup> we obtain

$$\alpha_N = \alpha nk / \text{Re}(\beta_N). \quad (\text{A6})$$

The difference between  $\alpha$  and  $\alpha_N$  is most substantial when a mode is near cutoff (*i.e.*  $kn \approx (N + 1)\pi/h$ ).

The rate constant  $\gamma_N$  for the dissipation of the areal energy density stored in a waveguide mode is obtained from the product of the attenuation constant and the group velocity

$$\gamma_N = \alpha_N v_g^N. \quad (\text{A7})$$

The group velocity of the guided mode is

$$v_g^N \equiv \left( \frac{d\beta}{d\omega} \right)^{-1} \cong \frac{\text{Re}(\beta_N)c}{n^2 k}. \quad (\text{A8})$$

We obtain by substitution of (A6) and (A8) into (A7)

$$\gamma_N = \alpha_N v_g^N = \alpha(c/n), \quad (\text{A9})$$

which is consistent with Eq. (A1).

It is interesting that the rate constant for energy decay in a guided mode is the same as its rate in radiative modes, independent of the group velocity of the mode. We think the relationship is sensible in terms of an underlying mechanism such as interband absorption, which is proportional to the local electromagnetic field energy density. For a given mode energy, the intensity of energy flow is of course proportional to the group velocity, which slows down compared to  $c/n$  when a mode is near its cutoff. This derivation indicates that the attenuation constant increases correspondingly, leaving the dissipation rate of the stored energy unchanged.

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