Tandem photonic-crystal thin films surpassing Lambertian light-trapping limit over broad bandwidth and angular range

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Abstract

Random surface texturing of an optically-thick film to increase the path length of scattered light rays, first proposed nearly thirty years ago, has thus far remained the most effective approach for photon absorption over the widest set of conditions. Here using recent advances in computational electrodynamics we describe a general strategy for the design of a silicon thin film applicable to photovoltaic cells based on a quasi-resonant approach to light trapping where two partially-disordered photonic-crystal slabs, stacked vertically on top of each other, have large absorption that surpasses the Lambertian limit over a broad bandwidth and angular range.

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One of the fundamental issues underlying the design of silicon photovoltaic (PV) cells for use in realistic settings is the maximum absorption of incident sunlight over the widest possible range of wavelengths, polarizations and angles using the thinnest material possible. While random or so-called Lambertian texturing of the surface to isotropically scatter incident light rays into a weakly-absorbing thick film so as to increase the optical path length, as shown in Fig. 1(a), has thus far remained the most effective approach for light trapping over a wideband spectrum [1, 2], recent thin-film nanostructured designs including photonic crystals [3] shown in Fig. 1(b) exploiting the more-complicated wave effects of photons have explored the possibility of superior performance [4–18] but have been mainly limited to narrow bandwidths, select polarizations or a restricted angular cone typical of delicate resonance-based phenomenon. The introduction of strong disorder, while improving robustness, nevertheless comes at the expense of light trapping relative to the unperturbed case [19, 20]. As a result, no proposal for a nanostructured silicon thin film capable of robust, super-Lambertian absorption over a large fraction of the solar spectrum has yet been made. In this work, we present a new approach to light trapping made possible by recent advances in computational electrodynamics [21] based on the quasi-resonant absorption of photons that combines the large absorption of optical resonances with the broadband and robust characteristics of disordered systems via a stacked arrangement of ordered PC slabs augmented with partial disorder that surpasses by a wide margin the performance of an idealized Lambertian scatterer over a broad spectrum and angular cone.

Our tandem design, consisting of the same silicon film structured in two different ways and stacked vertically as shown in Fig. 1(c), is the photonic analogue of the multi-junction cell that employs three or more different semiconductor films where the electronic bandgaps add complimentarily to obtain wideband absorption. Here we demonstrate the utility of a photonic approach, employing geometric structure alone, to enhance light trapping which also offers improved performance but does not incur the constraints and limitations of optimally combining multiple material-specific electronic bandgaps as well as the significant fabrication challenges and costs of epitaxially growing films with mismatched atomic lattices on top of one another. We outline a two-part design strategy based on first maximizing, with a few-parameter gradient-free topology optimization the number of resonant-absorption modes by using two crystalline-silicon PC slabs with a fixed total thickness of 1 µm stacked on top of each other such that their individual resonances add complimentarily over the
wideband spectrum; and then in the final step introducing a partial amount of disorder to both lattices to maximize the overall light trapping and boost robustness to go well beyond the Lambertian limit.

In our earlier work, we showed how individual resonant-absorption peaks of a thin-film PC slab can be broadened using partial disorder leading to an overall enhancement of the wideband absorption spectra \[19\]. To understand quantitatively why disorder increases broadband light trapping in a PC, we use coupled-mode theory to derive an analytical expression for a single absorption resonance (at a frequency of \(\omega_0\)) which has only one coupling channel for external light (a slight simplification which helps to make clear the role of disorder) in terms of the decay lifetimes (proportional to the quality factor) for radiation \((\tau_{\text{rad}})\) and absorption \((\tau_{\text{abs}})\) by the material \[3\]:

\[
A(\omega) = \frac{4}{\tau_{\text{rad}}\tau_{\text{abs}}} \left(\omega - \omega_0\right)^2 + \left(\frac{1}{\tau_{\text{rad}}} + \frac{1}{\tau_{\text{abs}}}\right)^2.
\]

Broadband absorption for a thin-film PC design, consisting of a collection of such individual Lorentzian peaks, necessitates that we consider the total area spanned by eq. (1) which is equivalent to its absorption cross section:

\[
\int_{-\infty}^{\infty} A(\omega) d\omega = \frac{4\pi}{\tau_{\text{rad}} + \tau_{\text{abs}}}.
\]

The effect of disorder is to reduce the peak height but more importantly to broaden the peak width (proportional to \(1/\tau_{\text{rad}}+1/\tau_{\text{abs}}\)) via primarily a decrease in \(\tau_{\text{rad}}\) which therefore leads to an overall increase in broadband absorption from eq. (2) (though \(\tau_{\text{abs}}\) also changes with disorder due to variations in the nature of the guided mode, the change is much less pronounced than \(\tau_{\text{rad}}\) mainly because the material absorption coefficient is fixed). Note that this analysis is only valid when coupling to a resonant Bloch mode which is why introducing too much disorder and eliminating the peaks altogether, thus transitioning to non-resonant Anderson-localized modes, results in sub-optimal light trapping \[19\].

We consider the absorption of solar radiation in the wavelength regime spanning 600nm to 1100nm in which silicon is poorly absorbing and thus weak coupling to resonant Bloch modes of the PC most apparent. The overall light-trapping efficiency of each design can be quantified relative to an ideal perfect absorber which has unity absorptivity over the wavelength interval of interest by assuming that each absorbed photon with energy greater than
the bandgap of silicon generates an exciton which contributes directly to the short-circuit current (this is equivalent to an internal quantum efficiency of 100%). This corresponds to the following definition of light-trapping efficiency:

\[
\frac{\int_{600\text{nm}}^{1100\text{nm}} \lambda I(\lambda) A(\lambda) d\lambda}{\int_{600\text{nm}}^{1100\text{nm}} \lambda I(\lambda) d\lambda},
\]

where \(I(\lambda)\) is the terrestrial power density per unit wavelength from the sun at AM1.5 [22] and \(A(\lambda)\) is the absorptivity of the film.

The design strategy of maximizing the light-trapping efficiency by controllably introducing a partial amount of disorder to obtain just the right dose of peak broadening ultimately results in a more-uniform absorption profile where the absorptivity in the inter-peak regions is increased at the expense of the height of all peaks. This therefore suggests that in order to most effectively make use of partial disorder for light-trapping enhancement in a thin film the number of resonant modes must first be made as large as possible so that there is little bandwidth separation between peaks: by extending our previous slab design to include not one but two PC lattices stacked on top of one other and separated by a non-absorbing nanoscale gap layer, such that the absorption spectra of each lattice when combined adds complimentarily (i.e., regions of low absorption in one lattice are compensated for by the high absorption of the other), the resonant-absorption characteristics of the PC augmented by partial disorder can potentially be exploited to the fullest extent possible for broadband absorption while also remaining feasible to large-scale industrial manufacturing. The low-index gap separation layer itself also provides additional mechanisms for light localization in nanostructured media that further contributes to enhancing absorption in the adjacent high-index layers while its effect on scattering-based Lambertian-textured films is marginal. A schematic of this design approach, somewhat exaggerated for illustrative purposes, is shown in Fig. 1(d) and (e) in which the tandem structure is first optimized for peak density in the spectrum (which amounts to maximizing the number of non-overlapping resonant absorption modes of the two constituent PC lattices) and subsequently these narrow closely-spaced resonances are slightly broadened with the addition of disorder to create a more-uniform absorption profile that is large in amplitude, broadband and robust to incident radiation conditions. We consider here for simplicity the case of two PC slabs in vacuum separated by an air gap which both incorporates all essential physical phenomena and has
direct applications to conventional thin-film PV cells in which individual layers including both high-index semiconductors and low-index transparent conductive oxides are grown by thin-film deposition tools \[23, 24\]. There is no need in the scope of the present work where the focus is solely on photon absorption in silicon to include an anti-reflection (AR) coating in the front or a perfect reflector in the back as would be customary in an actual PV cell since the role of both components is mainly to enhance, oftentimes significantly \[8, 25\], the absorption of existing resonances in the nanostructured films but not to give rise to new ones. Due to the complimentary way that the individual peaks of the two PCs combine in the tandem structure, a simple square lattice arrangement is adequate for good performance rendering unnecessary the need for intricate superlattice structures \[11, 18\].

To perform the topology optimization, we combine the capabilities of Meep, a freely-available open-source finite-difference time-domain (FDTD) tool \[26\], to compute the absorption spectra at normal incidence with the nonlinear-optimization routines of NLopt \[27\] (details in Supplementary Information). Here we need not consider absorption at off-normal incidence since the addition of disorder in the final step will automatically reduce the sensitivity to incident radiation conditions \[19\]. Accurate topology optimization is made possible using Meep’s subpixel averaging \[21, 26, 28\] which also significantly reduces the size of the computation by lowering the minimum spatial resolution required for reliable results. This is important since the objective function is iterated over a large number of times to explore small changes to geometrical parameters in order to engineer as many absorption resonances over the wideband spectrum as possible which tend to be highly susceptible to numerical artifacts introduced by “staircased” features \[21, 26, 28\] enabling FDTD to be used as a versatile 3D design optimization tool. We use intrinsic crystalline silicon as our absorbing material and incorporate its full broadband complex refractive index profile \[29\] into the FDTD simulations with accuracy even near its indirect bandgap of 1108nm where absorption is almost negligible to obtain experimentally-realistic results (see Supplementary Information for more details). Although, for generality, modeling a tandem structure consisting of two completely-independent PC slabs with arbitrary lattice constants is preferred, incorporating two distinct unit cells into a single 3D simulation is computationally impractical yet this is a minor design limitation as the other six structural parameters – as shown in Fig. \[1\](c): the thickness of the top Si layer \(v_t\) (the bottom thickness \(v_b\) is known since the total thickness is fixed at 1\(\mu\)m), the gap thickness \(g\), the radius and height of the holes
in the top and bottom lattices \( r_t, h_t, r_b, h_b \) – provide sufficient flexibility for creating out-coupled Bloch-mode resonances over the entire range of the broadband solar spectrum. We also investigated the computationally tractable case of two PC slabs with lattice constants differing by a factor of two though the results were not found to be an improvement to the single lattice-constant design. A planewave source is incident from above onto the tandem structure and the absorption spectrum \( A(\lambda) \), equivalent to 1-reflection-transmission, is calculated by Fourier-transforming the response to a short pulse. The absorptivity threshold used by the objective function to count the total number of peaks in the spectrum is taken to be that of our baseline performance metric: an equivalent 1\( \mu \)m-thick Si film with Lambertian-textured surfaces [2, 30] which has an efficiency of 43.0% in our wavelength interval (computed using the same fitted material parameters as used in the simulations). Since resonances with especially-large peak amplitudes contribute most to increasing the overall efficiency when broadened, we add an extra 30% to our absorptivity peak threshold at each wavelength which while making the problem more challenging gives rise to better results. We impose no restrictions on the peak width or spacing relative to other peaks although these could potentially be used for further refinement. Once an optimal set of parameters for the two-lattice structure is determined by running multiple times with different randomly-chosen initial values to explore various local optima, we then form a supercell consisting of 10x10 unit cells of the optimal structure and add positional disorder to each unperturbed hole in both lattices (while ensuring no overlap between holes to conserve the filling fraction) by an amount \( \Delta p_1 \) (\( \Delta p_2 \)) chosen randomly from a uniform distribution of values between 0 and \( \overline{\Delta p_1} \) (\( \overline{\Delta p_2} \)) for both orthogonal in-plane directions for the top (bottom) slab. Three separate simulations are made for each structure and the results averaged due to the random nature of the design.

Figure 2(a) shows the absorptivity spectra for three thin-film designs each with a total crystalline-silicon thickness of 1\( \mu \)m: an unpatterned film, a Lambertian-textured film [obtained from eq. (1) of Ref. 30] and finally the topology-optimized tandem design consisting of two PC slabs (top: thickness 708nm, hole radius 236nm, hole height 260nm and bottom: thickness 292nm, hole radius 199nm, hole height 244nm) with a lattice parameter of 641nm separated by a 228nm gap. The tandem design has numerous narrow, high-amplitude peaks – signatures of the coherent-resonant Bloch modes – that span the entire broadband spectrum whereas the unpatterned slab has broad Fabry-Pérot resonances with low amplitude.
The complimentary way that the resonances of the individual slabs combine to form the tandem structure can be seen in Figure 2(b) and (c) where the top slab accounts for most of the total number of peaks while the bottom slab contributes a few key resonances particularly at longer wavelengths. Note that while the Lambertian-textured slab has little and diminishing absorption at long wavelengths where the absorption coefficient of crystalline silicon is vanishingly small the PC design, due to its resonant nature, has large absorption albeit appearing only as very-narrow peaks (due to the rate-matching phenomena discussed previously that underlies the resonant coupling between radiation and absorption by the material, as silicon’s absorption coefficient becomes smaller at larger wavelengths leading to a corresponding increase in $\tau_{abs}$, the total lifetime of the resonant mode $\tau_{tot}$ being $1/\tau_{tot}=1/\tau_{abs}+1/\tau_{rad}$ also increases resulting in the inversely-proportional peak width generally becoming narrower which can be seen in Fig. 2). Nevertheless, the tandem design with its maximized peak density, outperforms the Lambertian texture in light-trapping efficiency (48.8% versus 43.0%) although at off-normal angles and different polarizations of incident light the unperturbed lattices’ resonance-based performance degrades significantly to below the Lambertian limit. Within the stacked arrangement, the efficiencies of the top and bottom slabs are 42.6% and 6.2%, respectively, while in isolation they are 37.9% and 22.3% highlighting in part the importance of inter-slab interactions to the overall absorption of the tandem design. For comparison, the optimized 1µm-thick single-slab design (lattice parameter, hole radius and height of 640nm, 256nm and 400nm, respectively) produces seven fewer resonances than the tandem design over the same wideband spectrum and therefore had a lower efficiency: nearly 5% below in absolute terms, yet still above the Lambertian limit.

By proceeding to controllably introduce a partial amount of disorder into the topology-optimized tandem design, we can simultaneously boost efficiency and improve robustness to exceed the Lambertian limit by an even wider margin over a large angular cone. Fig. 3(a) is a plot of the efficiency from eq. (3) versus disorder for the optimized tandem-slabs and single-slab designs at normal incidence and shows that a positional disorder of approximately $\Delta p_1=\Delta p_2=0.1a$ for the tandem slabs and $\Delta p=0.15a$ for the single slab results in maximal light trapping of 9.8% and 6.6% above the Lambertian limit, respectively, while additional disorder beyond these partial amounts leads to a steady decrease of the efficiency in line with the analysis presented earlier. The tandem design therefore is roughly twice as effective
as the single slab in overcoming the Lambertian limit due mostly to facilitating a larger number of absorption resonances. We quantify the performance robustness of each design as the standard deviation of the efficiency averaged over normal (0°) incidence and five off-normal (10°, 20°, 30°, 40°, 50°) angles of incidence for both S and P polarizations. A demonstration involving a larger angular range is possible but the necessary simulation times to ensure that the Fourier transforms used to compute the flux spectra have properly converged become prohibitively long. Since more disorder results in better robustness [19] which is a key requirement of a practical solar cell we make a slight trade off and apply not the quantities which maximize efficiency at normal incidence in the tandem design but values slightly greater ($\Delta p_1=0.2a$ and $\Delta p_2=0.25a$) where the robustness is substantially larger: 49.5%±2.3% for the former versus 49.4%±1.7% for the latter. Fig. 3(b) demonstrates that the average performance of this tandem design has greater absorption than the Lambertian texture at every wavelength resulting in a light-trapping improvement of almost 10% above the Lambertian limit.

In summary, we have described a general design strategy derived from a new conceptual framework of photon capture for a nanostructured silicon thin film based on the quasi-resonant absorption of photons in a tandem arrangement of partially-disordered photonic-crystal slabs separated by a nanoscale gap where the overall light trapping surpasses a Lambertian-textured film by a wide margin over a large fraction of the solar spectrum and a broad angular cone.

ACKNOWLEDGMENTS

This work was supported by Core Research for Evolutional Science and Technology (CREST) from the Japan Science and Technology Agency. A.O. was supported by a post-doctoral fellowship from the Japan Society for the Promotion of Science (JSPS).

AUTHOR CONTRIBUTIONS

A.O. conceived of the entire idea and performed all the simulations and analysis. A.O. discussed the results and wrote the manuscript with S.N. and Y.T. S.N. organized the project
and nurtured the environment for inquiry into how to increase absorption in thin films.

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FIG. 1. (a) Random or so-called Lambertian texturing of the surface to isotropically scatter incident light rays into the plane of a weakly-absorbing film so as to increase the optical path length. First proposed nearly thirty years ago, this has thus far remained the most effective approach for light trapping over the widest set of conditions. (b) Photonic-crystal slab and other nanostructured designs in which light trapping occurs by resonant absorption into a guided mode depend on delicate wave-interference effects and are thus intrinsically narrowband and restricted to a small angular cone. (c) Tandem arrangement of two PC slabs, both consisting of a square lattice of holes in silicon, stacked vertically on top of each other. Shown are the degrees of freedom – slab thicknesses $v_1$ and $v_2$, lattice parameters $a_1$ and $a_2$, hole radii $r_1$ and $r_2$, hole heights $h_1$ and $h_2$ and gap separation $g$ – used in the topology optimization to (d) engineer as many non-overlapping resonances over the wideband solar spectrum as possible. Following this, (e) each hole is perturbed from its position in the unperturbed lattice by amounts $\Delta p_1$ ($\Delta p_2$) chosen randomly from a uniform distribution of values between 0 and $\Delta p_1$ ($\Delta p_2$) for both orthogonal in-plane directions of the top (bottom) slab to boost light trapping and robustness by creating a more-uniform absorption profile.
FIG. 2. (a) Absorption versus wavelength profile at normal incidence for three thin-film PV designs each with a total crystalline-silicon thickness of 1 µm: an unpatterned slab (green), a Lambertian-textured slab (red) [obtained from eq. (1) of Ref. 30] and the topology-optimized tandem PC slabs (blue). The tandem PC slabs both consist of a square lattice (periodicity, $a=641$ nm) of holes in silicon separated by a 228 nm gap. Shown for each design is the photon-absorption efficiency defined in eq. (3) which is a measure of light trapping relative to a perfect absorber. (b) and (c) Individual absorption spectra for the top (slab #1: $v_t=708$ nm, $r_t=236$ nm, $h_t=260$ nm) and bottom (slab #2: $v_b=292$ nm, $r_b=199$ nm, $h_b=244$ nm) PC slabs of the optimized tandem design demonstrating how the resonances add complimentarily over the broadband spectrum.
FIG. 3. (a) Light-trapping efficiency from eq. (3) as computed from the absorption profile at normal incidence versus hole positional disorder for the topology-optimized tandem slabs (blue) and single slab (red) showing that partial disorder (tandem: $\Delta p_1 = \Delta p_2 = 0.1a$, single: $\Delta p = 0.15a$) maximizes the light trapping (tandem: 52.8%, single: 49.6%) while additional disorder is suboptimal and leads to a steady decline. Note that the tandem design is nearly twice as effective as the single-slab design in surpassing the Lambertian limit. (b) Absorption versus wavelength profile at normal (0°) incidence and five off-normal (10°, 20°, 30°, 40°, 50°) angles of incidence averaged over both $S$ and $P$ polarizations of the optimized tandem design with the addition of partial disorder of $\Delta p_1 = 0.2a$ and $\Delta p_2 = 0.25a$ as shown in the inset schematics. The absorption profile for the individual angles are colored while the average of all the data is shown in black which exceeds the Lambertian texture over the entire interval resulting in an overall light-trapping efficiency that is approximately 10% greater.