Thermodynamic Upper Bound on Broadband Light Coupling with Photonic Structures

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Phys. Rev. Lett. **109**, 173901 — Published 23 October 2012

DOI: 10.1103/PhysRevLett.109.173901

Thermodynamic upper bound on broadband light coupling with photonic structures

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The coupling between free space radiation and optical media critically influences the performances of optical devices. We show that for any given photonic structure, the sum of the external coupling rates for all its optical modes are subject to an upper bound dictated by the second law of thermodynamics. Such bound limits how efficient light can be coupled to any photonic structure. As one example of application, we use this upper bound to derive the limit of light absorption in broadband solar absorbers.

Optical structures are fundamentally characterized by the modes that they support. For a closed and lossless structure, a mode is characterized by a steady state solution of Maxwell's equations at a single frequency ω. When such a structure is coupled to external radiation and also subject to internal loss, each mode is additionally described by an external coupling rate γ and internal loss rate $\tilde{\gamma}$ [1]. Rates are defined as the total energy stored in the structure divided by the power loss due to either external radiation or intrinsic loss.

Many practical optical devices that are related to energy applications, such as solar cells, light emitting diodes, and thermal radiators, are inherently large-area devices with a device size that is much larger than the wavelength of light. (Here we would like to make a distinction between device area, which is typically large, and the minimum feature size, which in our work can be at single- or even deep subwavelength scale). Moreover, the operating spectral bandwidth of these devices is typically quite broad. It is typically on the order of kT, where T is the sun's temperature for a solar cell, or the operating temperature for a thermal radiator. In such situations, to understand device performance it is not sufficient to characterize

the property of only a single optical mode. Instead, one needs to understand the statistical properties of a collection of modes.

In this paper we show that for all modes in a given frequency range $[\omega, \omega+\Delta\omega]$, the sum of their external coupling rate is subject to an upper bound. We prove the existence of this bound using the second law of thermodynamics. This bound establishes a fundamental constraint on how efficiently broadband radiation can couple in and out of a large-area structure, and is therefore important for a wide range of devices, including thermal radiators, solar cells, and light emitting diodes. As an application, we show that one can use this bound to derive the limit of light trapping enhancement for nanophotonic solar cells, even in cases where the single-pass absorption of the cell is non-negligible. This is in contrast with previous theoretical works on nanophotonic light trapping that have focused only on the infinitesimal absorption limit[2].

We begin by considering a slab of a medium with arbitrary structure and with a large area L^2, where L is the side length of the structure (Fig. 1). To facilitate our theoretical treatment, we use periodic boundary conditions on the edges of the structure. We further assume that the slab is placed on a perfectly reflecting surface, such that the modes in the slab only couple to free space through the top surface. For optical modes in the frequency range between ω and ω+Δω, the sum of their external coupling rates for this structure is subject to the following upper limit:

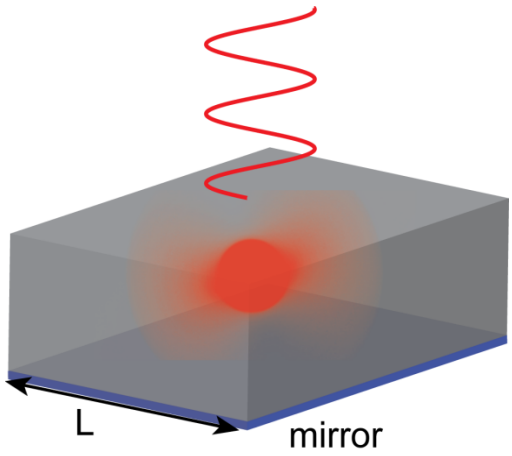

Fig. 1. Schematic of an optical resonance coupling into free space through the top surface.

$$\sum_m \gamma_m \le \frac{L^2\omega^2}{4\pi^2 c^2}\Delta\omega \qquad \text{Eq. (1)}$$

where c is the speed of the light in vacuum. m labels different mode. If we further assume an equal coupling rate $\gamma_m = \gamma$ for all modes, Eq. (1) becomes

$$\gamma \le \frac{c}{4d}\frac{\rho_{vac}}{\rho} \qquad \text{Eq. (2)}$$

where ρ is the optical density of states (DOS) in the medium. $\rho_{vac} = \frac{\omega^2}{\pi^2 c^3}$ is the DOS in vacuum. d is the thickness of the medium.

To derive Eq. (1), we first assume that the medium has infinitesimally small absorption, and is in thermal equilibrium with its environment. The number of photons in each optical mode in the frequency interval $[\omega, \omega+\Delta\omega]$ is $1/(e^{\hbar\omega/k_B T}-1)$, where k_B is the Boltzmann constant and T is the temperature. Since each such optical mode radiates photons to free space with external coupling rate γ_m, the number of emitted photons per second from each such mode is

$$p_m = \frac{\gamma_m}{e^{\hbar\omega/k_B T}-1} \qquad \text{Eq. (3)}$$

On the other hand, according to Planck's law, a black body at the same temperature with the same device area L^2 emits a total photon flux

$$B = L^2 \frac{\omega^2}{4\pi^2 c^2}\frac{1}{e^{\hbar\omega/k_B T}-1}\Delta\omega \qquad \text{Eq. (4)}$$

By the second law of thermodynamics, more specifically, the Kirchhoff's law, the total emission from the slab cannot be larger than that from the blackbody at the same temperature, hence:

$$\sum_m p_m \le B \qquad \text{Eq. (5)}$$

which leads to Eq. (1).

We can further find the constraint that limits the total coupling rates to any particular spatial direction. The second law of thermodynamics limits the flux density that any thermal body in equilibrium can emit to each radiation channel in free space. For the geometry in Fig. 1, we define a radiation channel as characterized by a distinct wavevector $k_{\|}$ parallel to the surface of the emitter, and with one of the two polarizations associated with this direction. We use $\gamma_{m,n}$ to describe the coupling between resonance *m* and channel *n*. Thus, the total external coupling rate $\gamma_m = \sum_n \gamma_{m,n}$. If the emitter in Fig. 1 is a blackbody, its emitted flux to the *n*-th channel is [3-4]:

$$b_n = \frac{1}{e^{\hbar\omega/k_B T} - 1} \frac{d\omega}{dk_\perp} \frac{dk_\perp}{2\pi} = \frac{1}{e^{\hbar\omega/k_B T} - 1} \frac{\Delta\omega}{2\pi} \qquad \text{Eq. (6)}$$

For a general emitter, since total photons emitting to each channel cannot be greater than b_n , similar to the derivation of Eq. (1), here we have

$$\sum_m \gamma_{m,n} \le \frac{\Delta\omega}{2\pi} \qquad \text{Eq. (7)}$$

The summation in Eq. (7) is again for all optical modes in the slab with frequencies between ω and $\omega + \Delta\omega$.

Eqs. (1) and (7), which are the main results of this paper, provide fundamental constraints on the external coupling rates of optical modes to free space. While we have used the assumption of thermal equilibrium

to establish these upper bounds, these bounds themselves are valid no matter whether the slab is in thermal equilibrium or not. As one application, we will use Eqs. (1) and (7) to study light absorption in solar cells. In a flat cell, due to total internal reflection, a significant fraction of the optical modes in the cell do not couple to external radiation, and hence do not contribute to light absorption. The concept of light trapping in solar cells aims to use all optical modes in the cell and ensure that they couple to external radiation efficiently[2]. For this purpose, photonic structures, including surface texturing[5-11] in standard crystalline silicon solar cells, and more recently a wide variety of nanophotonic structures[10, 12-21], have been developed. Eqs. (1) and (7) show that for all these structures, there is a fundamental upper bound on how efficiently light can couple into them. By using this upper bound, one can obtain an upper limit on light-trapping absorption enhancement, including the case where the single pass absorption in the solar cell is non-negligible.

The connection, between the sum of the external coupling rates of all modes, with the maximum achievable absorption of broadband light, was established in the statistical coupled mode theory formalism of Ref. [2]. In this formalism, the total light absorption is described as the aggregate contribution of all optical resonances in the absorber. For a single optical resonance, its absorption spectrum $A_m(\omega)$ has a Lorentzian lineshape (See derivation in the Supplementary Information)

$$A_m(\omega) = \frac{\gamma_m \tilde{\gamma}_m / N}{(\omega - \omega_m)^2 + \left(\gamma_m + \tilde{\gamma}_m\right)^2 / 4} \qquad \text{Eq. (8)}$$

Here $\tilde{\gamma}_m$ is the absorption rate of the mode. ω_m is the resonance frequency. Light is incident from the n_{th} channel. The absorption $A_m(\omega)$ does not depend on the incident channel since we have further assumed that the resonance couples to N free space channels equally with $\gamma_{m,n} = \frac{\gamma_m}{N}$. In the cases where there is a large number of channels, equal coupling to these channels is equivalent to the assumption of the cell being a Lambertian emitter, an assumption that is commonly made in solar cell theory[6, 8]. Our theory

here is more general since it can be applied as well to systems with small number of channels such as a grating structure with wavelength scale periodicity. In the weak absorption limit, the general case for where coupling rates to different channels are not the same can be found in [22]. We believe similar generalization can be made in the non-negligible absorption regime as well.

The contribution of a single resonance to broadband absorption is characterized by its spectral absorption cross section. The absorption coefficient *A* is then calculated from the summation of the spectral absorption cross sections of all resonances within a frequency range $\Delta\omega$ (See more details in the supplementary information)

$$A = \frac{1}{\Delta\omega}\sum_m \int_{-\infty}^{\infty} A_m(\omega) d\omega = \frac{2\pi}{\Delta\omega}\frac{1}{N}\sum_m \frac{\gamma_m \tilde{\gamma}_m}{\gamma_m + \tilde{\gamma}_m} \qquad \text{Eq. (9)}$$

In deriving Eq. (9), we assume that the bandwidths of resonances are narrower than $\Delta\omega$ and thus the range of integration can be extended to infinity.

Eq. 9 shows that the absorption of the structure *A* increases when coupling rates γ_m increase. However, our results of Eq. (1) and (7) indicate that the coupling rates are upper-bounded, which in turn imposes a constraint on the maximum value of *A*. To simplify the algebra, we suppose that within the frequency interval of $[\omega,\ \omega+\Delta\omega]$ the material absorption is a constant. We further assume that all modes within such interval have the same absorption rate, i.e. $\tilde{\gamma}_m = \tilde{\gamma} = \alpha c / n$, where α is the absorption coefficient of the material and *n* is its refractive index. It can be shown using the method of Lagrange multipliers (See Supplementary Material) that *A* is maximized when all coupling rates γ_m are equal. From Eq. (7), we thus have $\gamma_m \le \frac{N}{M}\frac{\Delta\omega}{2\pi}$, where *M* is the total number of modes in the frequency range $[\omega, \omega+\Delta\omega]$. Substituting this limit into Eq. (9), we obtain (see details in supplementary material)

$$A \le \frac{\alpha d}{\alpha d + \frac{1}{F}}, \qquad \text{Eq. (10)}$$

where

$$F = \frac{M}{N}\frac{2\pi\tilde{\gamma}}{\alpha d \Delta\omega} \qquad \text{Eq. (11)}$$

is the maximum light trapping enhancement factor in the limit where the single pass absorption is infinitesimal, as derived in Ref.[2].

Eq. (10) is consistent with, and generalizes, previous results on light trapping absorption enhancement. For bulk media, F as determined from Eq. (11) is $4n^2$ assuming an isotropic emission pattern, as shown in Ref. [2]. Eq. 10 then reproduces the well-known Lambertian limit[5-6, 8, 23]

$$A_{Bulk} \le \frac{\alpha d}{\alpha d + 1/4n^2}. \qquad \text{Eq. (12)}$$

We will refer to this limit as the conventional limit for the rest of the paper.

When the period L is large, the number of modes M and the number of channels N can be calculated by assuming that these modes or channels form continuums. In this case, we can show that

$$F = \frac{4}{n}\frac{\rho}{\rho_{vac}} \qquad \text{Eq.(13)}$$

And the light absorption limit from Eq. (10) is

$$A \le \frac{\alpha d}{\alpha d + \frac{n}{4}\frac{\rho_{vac}}{\rho}} \qquad \text{Eq.(14)}$$

This result is consistent with Stuart and Hall[24]'s result for absorption limit in thin-film waveguides and recent work by Callahan, Munday and Atwater[25].

Our results, however, are more general. Eq. (10) is applicable even for structures where the number of resonances or the number of channels becomes discrete. Examples of such structures include gratings with periodicity comparable to the wavelength of the interest[26]. In this regime, the radiance theorem used in Ref [24] , which assumed a continuum of external radiation channels, is not easily applicable, and Eq. (13) no longer applies[24]. In addition, in deriving Eq. (10), we prove rigorously the connection between the regimes of infinitesimal material absorption, and the regimes of non-negligible material absorption. This connection indicates that nanophotonic structures can be used to overcome the conventional limit of broadband absorption enhancement for non-negligible material absorption strength.

We end the theory section with a brief comment on the regime of validity of some of the main results for light trapping. It has been noted that the conventional theory of Eq. (12) is applicable when $A < 0.9$[27]. Our theory, as a generalization of the conventional theory of Ref. 27, assumes the same regime of validity, i.e. our theory is applicable when the material absorption strength is non-negligible. This is in contrast with Ref. [2] where only the case of infinitesimal material absorption is considered. Our theory, similar to conventional light trapping theory, is not applicable in principle in the strong absorption limit of $\alpha d >> 1$. However, in practice there is essentially no interest to perform light trapping in such strong absorption limit since the single pass absorption is already sufficiently large. In this sense, our theory is therefore applicable for all regimes where light trapping is important.

As an application of the theory above we examine the structure shown in Fig. 2a. The absorber consists of a d=5nm thick active layer with frequency-independent absorption coefficient α . The active layer is surrounded by cladding layers with a high refractive index. For such a structure, the TE modes has a strong field concentration, and hence a greatly enhanced local density of state, at the active layer, due to the slot waveguide effect[28]. The structure has a multi-layer anti-reflection coating on top of cladding

layer. A scattering layer is placed on the back of the cladding layer to ensure efficient coupling to all modes. In contrast to the structure in [2], which consists of a single scattering layer on top of the structure, here we introduce an additional anti-reflection layer on top of the structure, and place the scattering layer at the bottom of the structure. In Ref. [2], it was shown that a single scattering layer is sufficient to achieve very large absorption enhancement factor in the weak absorption regime. In contrast, we have found here that to achieve substantial enhancement when the material absorption is stronger, the presence of the anti-reflection layer is essential. Also, since the structural requirement for anti-reflection and for scattering is different, the placement of these layers on the opposite side of the absorber enables separate optimization of these layers for these two different purposes (Fig. 2a)[29].

In the weak absorption regime, the conventional limit for bulk active materials is $F_{bulk} = 4n^2 = 10$. Due to the nanoscale confinement of TE modes in the active layer, the upper limit of absorption enhancement, in the regime where the material absorption is negligible, is $F = 200$ as calculated using the approach outlined in Ref. [2]. With this enhancement factor calculated, Eq. (10) allows us to examine the absorption limit outside the weakly absorptive regime. As an example, at $\alpha d = 0.02$, the upper limit of absorption is 80% for the nanostructure while it is 16.7% for bulk structures subject to the conventional limit.

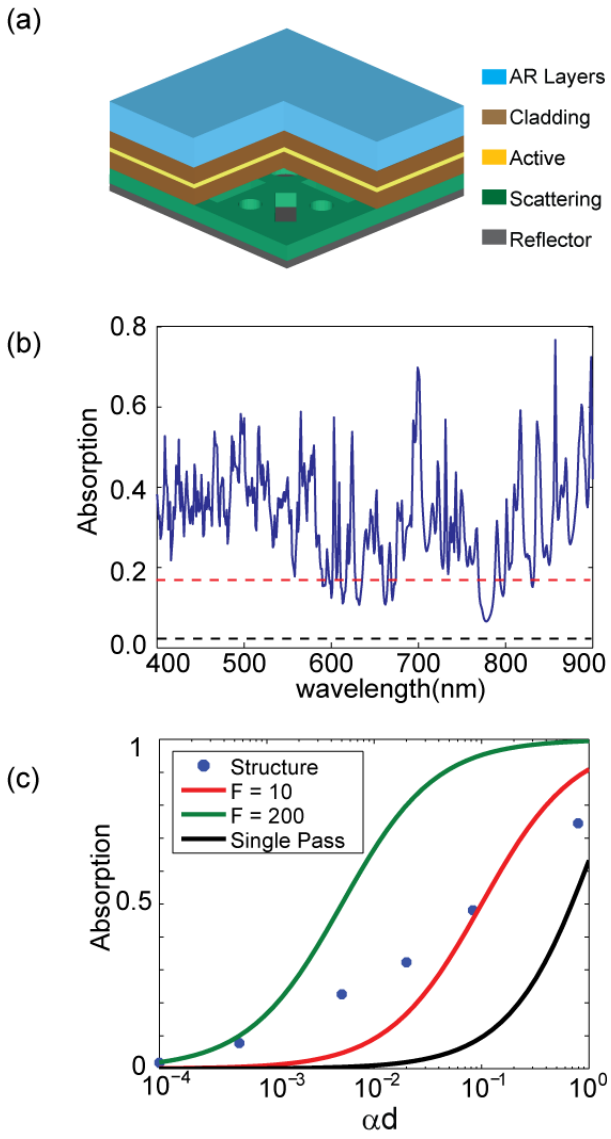

Fig. 2 a) Solar absorber structure (with cut-out showing the scattering layer pattern). It consists of cladding layers (brown color) of thickness 125 nm and ε= 12.5 surrounding the 5 nm active layer of ε = 2.5 on both sides. Below the lower cladding layer, a 150 nm thick scattering layer (green color) of air patterns is etched into a slab of ε = 12.5 with 1200 nm periodicity. The air patterns consist of eight rectangles of 200 x 50 nm radially separated at equal angles. In between each rectangle an air circle of radius 60 nm is also placed. Together they generate a strongly scattering layer to couple into desirable optical modes that are confined in the active layer. Below the scattering layer we placed a perfectly reflecting substrate. Finally, on top of the top cladding layer we introduce an optimized four layer anti-reflection (AR) coating. The layers have heights (from top to bottom) of 60 nm, 35 nm, 110 nm and 50 nm, and epsilons of 1.9, 3.7, 8.3 and 11.5 respectively. b)The absorption spectrum for single pass absorption of $\alpha d = 0.02$. This is in the parameter regime where material absorption is significant. The peaks are due to optical resonances. The absorption spectrum is above both the single pass absorption(black dashed line) and the conventional limit (red dashed line). c). The upper limit of light absorption A as the function of αd for absorbers for different F values calculated from Eq. (11). The black curve is for single pass absorption. Dots are simulated results for the structure. F = 10 is the conventional limit and F = 200 is the limit for the structure in a).

Next, we use numerical simulations to calculate the light absorption in the nanostructure shown in Fig. 2a. We use the RCWA method[30] with 22x22 Fourier orders to solve Maxwell's equations. In the weakly absorptive regime, for example $\alpha d = 0.0001$, the enhancement factor averaging from 400nm to 900nm wavelength is *F*=180, which agrees well with the analytical upper limit prediction of $F_{nano} = 200$, and is well above the conventional limit of 10. We then gradually increase the single pass absorption. For example, Fig. 2b shows the absorption spectrum when $\alpha d = 0.02$. The spectrally averaged absorption is 32%, or 16 times the single pass absorption, which is much higher than the conventional limit of 16.7% calculated from Eq. (12).

Simulation results for different absorption strengths are shown as dots in Fig. 2c, which follow the trend of the theoretical predictions as calculated by Eq.(10). In the regime of relatively weak single-pass absorption, the simulated absorption is very close to our theoretical upper limit. As the single pass absorption further increases, the simulated absorption starts to fall below our theoretical upper limit. This is expected. Optimal absorption is achieved when the external radiation rate dominates the intrinsic loss rate for all modes. As the material absorption increases, the intrinsic loss rate for all modes become larger, and it becomes progressively more difficult for the scatterers to function effectively. Nevertheless, the simulated absorption value for our structure (blue dots in Fig. 2c) is above the conventional limit (red curve in Fig. 2c), even for a relatively strong absorption coefficient of $\alpha d = 0.1$.

Our simulations thus directly demonstrate that with appropriate nanophotonic design, one can achieve absorption enhancement that is significantly beyond the conventional limit, even in the regime where the single-pass absorption is significant. Certainly, having a high local density of states in the absorbing layer is important for such absorption enhancement[2, 31]. In the mean time, however, it is also of critical importance to achieve maximum light coupling rates. This is particularly important when the single pass absorption is significant. The maximum coupling rates, corresponding to the equal sign in Eq. (1), in fact can be achieved only when the absorber has perfect anti-reflection properties. Optimal light trapping requires optimal anti-reflection properties. Lastly, if all resonances have similar intrinsic loss rates, it is

beneficial that these resonances have similar external coupling rates as well. The numerical results of Fig. 2 indicate that these theoretical considerations can be met with nanophotonic structures.

In conclusion, using the second law of thermodynamics, we have derived a thermodynamic upper bound to the coupling between broad-band light and the optical modes in arbitrary structures. This bound has important implications on the performance of a range of optical devices for energy and thermal applications. As an example, we use this upper bound to derive the upper limit of light absorption in solar absorbers. The combination of broadband DOS enhancement and efficient light coupling can lead to extraordinarily high absorption well beyond the conventional limit. Our formalism here provides a theoretical foundation to understand light absorption in solar cells, especially ultra-thin solar cells where wave effects are dominant [2, 32-33].

Acknowledgement

This work was supported in part by the Center for Advanced Molecular Photovoltaics (Award KUSC1-015-21), made by King Abdullah University of Science and Technology, and by Department of Energy Grant DE-FG02-07ER46426. A.R. also acknowledges the support of Alberta Scholarship Programs' Sir James Lougheed Award of Distinction.

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