Broadband enhancement of light harvesting in luminescent solar concentrator

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Luminescent solar concentrators (LSCs) are large-area devices that may absorb incident sunlight, then emit luminescence photons with high quantum efficiency which finally be collected by a small photovoltaic (PV) system. The light-harvesting area of the PV system is much smaller than that of the LSC system, potentially reducing the cost of solar cells. Here, we present a theoretical description of the luminescent process in nanoscale LSCs where the conventional ray-optics model is no longer applicable. We demonstrate that a slot waveguide consisting of a nanometer-sized low-index slot region sandwiched by two high-index regions provides a broadband enhancement of light harvesting by the luminescent centers in the slot region. This is because the slot waveguide can (1) greatly enhance the spontaneous emission due to the Purcell effect, (2) dramatically increase the effective absorption length of luminescent centers, and (3) strongly improve the fluorescence quantum yield of luminescent centers. It is found that about 80% solar photons can be re-emitted even for a low fluorescent quantum yield of 0.5, and 80% re-emitted photons can be coupled to the slot-waveguide. This LSC is potential to construct a tandem structure which can absorb nearly full-spectrum solar photons, and also may be of special interest for building integrated nano-solar-cell applications.

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In the past few years, many approaches involving nanostructures or nanostructured materials have been proposed to reduce cost and improve efficiency in both experiment and theory. On the other hand, concentrators with large-area optical components to collect direct sunlight and transfer the energy to small, high-efficiency photovoltaic (PV) cells have been suggested as a simple approach to lower the cost per peak Watt of solar cell systems for many decades. To overcome the excess heat problem, chromatic aberrations and expensive maintaining in these imaging concentrators, luminescent solar concentrators (LSCs) represent an alternative approach to lower the costs of solar cell systems. LSCs generally consist of low-cost transparent sheets doped with luminescent species, such as dye molecules and quantum dots. Incident sunlight is highly absorbed by the luminescent centers and luminescence is emitted with high fluorescence quantum yield (FQY, defined as the ratio of the number of photons emitted to the number of photons absorbed), so that the emitted photon is trapped in the sheet by total internal reflection and travels to the edges where it can be collected by solar cells. The active material layer can be much thinner than the intrinsic absorption length of the material, thus dramatically reducing the amount of the solar cell material.

One of the key parameters of LSCs is the coupling efficiency $\beta$ which describes the ratio of luminescent photons coupled to waveguide modes for ultimate collection by PV systems on the waveguide edge. To model these LSCs devices, thermodynamical and computational ray tracing approaches have been introduced, and both approaches represent a broad-scale, macroscopic model. For example, conventional LSCs have a thickness much larger than the wavelength, so that the ray-optics model can be applied to estimate the collection efficiency of luminescence by the solar cell. However, with LSCs devices moving to the nanoscale, the ray-optics picture and some basic assumptions are no longer strictly applicable. In this paper, we propose a nanometer-sized slot waveguide as the main structure of LSCs, and theoretically study the enhancement of spontaneous emission (described by the factor $F_{p}$) of luminescent centers in this slot waveguide based on the Fermi-golden rule. Remarkably, this great enhancement of spontaneous emission predicts not only an increased absorption length of luminescent centers but also a very large waveguide coupling efficiency $\beta$. We demonstrate that such a slot waveguide LSC provides a broadband enhancement of light harvesting. It is found that about 80% solar photons can be re-emitted even for a low initial FQY of 0.5, and 80% re-emitted photons can be coupled into the slot-waveguide modes for ultimate collecting by the solar cell located on the waveguide edge. This LSCs may be of special interest for building integrated solar-cell applications.

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FIG. 1: Schematic illustration of a slot waveguide structured LSC. The slot waveguide consists of a nanometer-sized low-index slot region sandwiched by two high-index regions. Active medium such as dye molecules and quantum dots are located in the slot region. The PV system is on the edge of the slot waveguide and can collect the luminescent photons coupled in the slot waveguiding mode.
The basic structure of the proposed design is shown in Figure 1(b). The slot waveguide consists of a nanometer-sized low-index slot region (with the permittivity $\varepsilon_1 = n_1^2$, here $n_1$ is assumed 1.4) sandwiched by two high-index transparent layers (with the permittivity $\varepsilon_2 = n_2^2$, here $n_2$ is assumed 2.4), and the PV systems are on the edge of the slot waveguide. In this work, high-index polymer or glass layers are desirable to indeed reduce the cost, but it requires future studies. Active medium such as dye molecules and quantum dots are attractive for numerous applications, for instance, highly sensitive biosensors [19] and waveguide-based light source [20]. In this paper, also due to the large electric field intensity of the slot waveguide mode, the luminescent photons are expected to couple to the propagating slot mode, and finally be collected by the smaller high-efficiency PV systems. Due to the concentration effect at the LSCs edges, the amount of the solar cell material reduces dramatically, and the cost of solar cell may decrease greatly with the help of the slot waveguide LSC. To analyze this LSC system, we note that the conventional ray-optics model is not applicable because the thickness of the slot region is nanometer-sized. Thus, we derive a simple analytical formula from the Fermi-golden rule to obtain the spontaneous emission enhancement $F_p$ and the waveguide coupling ratio $\beta$. In the weak-coupling regime, the spontaneous emission rate of a dipole can be calculated from

$$\gamma_{WG} = 2\pi |g(\vec{r})|^2 \rho(\omega),$$

(1)

where $|g(\vec{r})|$ denotes the coupling strength between the dipole $\vec{d}$ and the electromagnetic field $\vec{E}$ at the dipole position $\vec{r}$. $\rho(\omega)$ represents the density of states. If we assume that the dipole is oriented parallel to the electric field, the coupling strength $d$ is oriented parallel to the electric field, the coupling strength $\beta$ is oriented parallel to the electric field, the coupling strength $\rho(\omega)$ is oriented parallel to the electric field, the coupling strength

$$\gamma_{WG} = 2\pi |d|^2 W(\vec{r}) \frac{\omega}{2\hbar \varepsilon_0 \varepsilon_1 A_{eff} \pi v_g(\omega)}.$$

(5)

is then given by

$$|g(\vec{r})| = |\vec{d} \cdot \vec{E}(\vec{r})/\hbar|,$$

(2)

with

$$\vec{E}(\vec{r}) = \sqrt{\frac{\hbar \omega}{2 \varepsilon_0 \varepsilon_1 A_{eff}}} W(\vec{r}) \frac{\vec{E}(\vec{r})}{|\vec{E}(\vec{r})|}.$$

(3)

Here $W(\vec{r})$ designates the normalized electromagnetic energy density distribution; $A_{eff}$ defines the effective mode area as

$$A_{eff} = \iiint_V \varepsilon_0 \varepsilon_1 \left| \vec{E}(\vec{r}) \right|^2 \, dV / \max \left[ \varepsilon_0 \varepsilon_1 \left| \vec{E}(\vec{r}) \right|^2 \right],$$

which plays the role analogous to the mode volume in cavity QED; $l$ is an arbitrary quantization length which can be canceled later. For a slot waveguide with a sufficiently small cross-sectional area, it only supports a single quasi-TE and a single quasi-TM mode. As mentioned before, the TE mode has the maximum electric field in the slot region. Thus, assuming a one-dimensional density of states, the density of states can be expressed as

$$\rho(\omega) = \frac{l}{\pi v_g(\omega)},$$

(4)

where $v_g = c/n_g$ is the group velocity of the slot waveguide mode with a group index $n_g$. Here, $c$ is the light velocity in vacuum. Combining Equations (1)-(4), we can obtain the spontaneous emission rate of the dipole in the slot waveguide

FIG. 2: (a), (b) False-color representations of electromagnetic energy density distributions $W(\vec{r}) = \varepsilon_0 \varepsilon_1 (\vec{E}(\vec{r}) \omega) d\omega/\varepsilon_0 \varepsilon_1 |\vec{E}(\vec{r})|^2 + \mu_0 |\vec{H}(\vec{r})|^2$ for quasi-TM and -TE modes, respectively, where the red arrows show the directions of the electric fields. The geometry of the slot waveguide is depicted in (b). (c), (d) Distributions of the normalized energy density $W(\vec{r})$ along the y direction at several typical x positions for quasi-TM and -TE modes, respectively. Here, $\lambda = 700$ nm; $a = 100$ nm, $b = 150$ nm and $g = 10$ nm; $n_1 = 1.4$, $n_2 = 2.4$. 

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(5)
With the spontaneous emission rate into free space $\gamma_0 = d^2 \omega / (3 \pi \hbar c^3)$, the emission enhancement also known as Purcell effect, is thus expressed as

$$F_p \equiv \frac{\gamma_{\text{WG}}}{\gamma_0} = \frac{3}{4\pi} \frac{c}{v_g(\omega)} \left( \frac{\lambda_0/n_1}{\lambda_0} \right)^2 W(\bar{r}). \quad (6)$$

Clearly, if the effective mode area $A_{\text{eff}}$ of the slot waveguide mode is squeezed significantly below $\lambda_0^2$, the light-matter interaction can be dramatically enhanced, thus leading to an enhanced $F_p$. In addition, the group velocity in the slot waveguiding mode is reduced, which can furthermore increase the local density of states and also contribute to the increase of $F_p$. Once the emission enhancement is given, the waveguide coupling efficiency $\beta$ describing the ratio of the emission coupled to the slot waveguiding mode, can be calculated as

$$\beta = \frac{F_p}{F_p + n_1}. \quad (7)$$

Equations (6) and (7) represent the main physical parameters of the proposed LSC. To numerically evaluate the emission enhancement and the waveguide coupling efficiency, we still resort to the FEM simulation, as it can provide not only the mode distribution ($f(\bar{r})$, $A_{\text{eff}}$) but also the group velocity $v_g(\omega)$ for a given geometry and working wavelength. Figure 3(a) shows the calculated $F_p$, and $\beta$ as a function of free-space wavelength $\lambda_0$ when a dipole is placed in the center of the cross-section of the slot waveguide. We find two points. (i) $F_p$ first increases and then decreases with the emission wavelength from 500 to 900 nm. This phenomenon is because that the effective mode area $A_{\text{eff}}/\lambda_0^2$ first increases and then decreases. For the short wavelength, most energy distributes in the high-index layers, and the field enhancement effect in the slot becomes weaker. As a result, the effective mode area increases. For the long wavelength, the small size of the slot waveguide cannot support the guiding mode well. Therefore, more and more energy diffuses outside for longer wavelength, and the effective mode area strongly expands. (ii) $F_p$ exceeds 7 over a broad wavelength range (500 – 800 nm). For longer wavelengths, a larger-sized slot waveguide can be employed to reach the maximum $F_p$, which will be discussed in the following. This strong enhancement of the spontaneous emission takes advantage of nonresonance (large bandwidth), which benefits from the subwavelength mode area (as small as 0.02 ($\lambda_0/n_1)^2$) and the reduced group velocity (with the group index $n_g \sim 2$ – 2.7) provided by the slot waveguide. Remarkably, this broadband enhancement character overcomes the narrow bandwidth limit of a microresonator-based emission enhancement. As a result, most of the spontaneous emission can efficiently couple to the slot waveguide mode, and $\beta$ exceed 0.8 over a broad wavelength range (500 – 800 nm).

The enhancement of spontaneous emission also depends on the geometry of the slot waveguide, for example, the slot width. Figure 3(b) shows the calculated enhancement factor $F_p$ as a function of the slot width $g$. The factor $F_p$ increases monotonically with the slot width reducing. For example, $F_p$ increases from 5 to 14 when $g$ decreases from 50 to 5 nm. The underlying physics is that both the effective mode area and the group velocity are strongly reduced by decreasing the slot width. For instance, $A_{\text{eff}}$ and $n_g$ are 0.0223 $\mu m^2$ and 2.15 at $g = 50$ nm, 0.0104 $\mu m^2$ and 2.82 at $g = 5$ nm. Thus, in this slot width range, the waveguide coupling efficiency $\beta$ changes from 0.78 to 0.9.

As mentioned above, we use 2.4 as the default $n_2$. However, for a different $n_2$, $F_p$ and $\beta$ can experience a significant change. From Figure 3(c) we can see that with a larger $n_2$, both $F_p$ and $\beta$ improve, which is because high-index-contrast interfaces can bring out a better enhancement and confinement of light [18]. Therefore, materials with both high transparency and refractive index are strongly preferred in our work.

In the analysis above, we have assumed that the dipole is
oriented parallel to the electric field of the quasi-TE mode. In an actual case, the orientation of a dipole may be isotropically distributed over any direction. In general, the quasi-TE mode has a much stronger electric field concentration in the slot region than the quasi-TM mode. As a result, the dipole oscillating in the \( y \) direction (coupled to the quasi-TE mode) exhibits a much larger emission enhancement than the dipole oscillating in the \( x \) direction (coupled to the quasi-TM mode). In addition, the spontaneous emission enhancement of the dipole oscillating in the \( z \) direction is even smaller because both the modes are quasi-transverse. The isotropically averaged emission enhancement factor \( \bar{F}_p \) can be calculated by

\[
\bar{F}_p = \frac{F_{p,x} + F_{p,y} + F_{p,z}}{3}.
\]

where \( F_{p,i=x,y,z} \) denotes the spontaneous emission enhancement when the dipole oscillates in the \( i \) direction. With \( \bar{F}_p \), the isotropically averaged waveguide coupling efficiency \( \bar{\beta} \) is thus obtained as

\[
\bar{\beta} = \frac{\bar{F}_p}{\bar{F}_p + n_1}.
\]

Figures 4(a) and 4(b) show \( \bar{F}_p \) and \( \bar{\beta} \), respectively, depending on the emission wavelength for three different slot widths \( g = 10, 20, 30 \) nm. From Figure 4(a), on one hand, due to enhanced dipole-field coupling, \( \bar{F}_p \) increases as the slot width decreases. On the other hand, \( \bar{F}_p \) still keeps high over the whole calculation range from 500 to 800 nm. Thus, even if dipoles oscillate in random directions, most emission of the dipoles will couple into the waveguide modes. For instance, as demonstrated in Figure 4(b), the averaged waveguide coupling efficiency \( \bar{\beta} \) is high above 0.7 over a broad wavelength range in the case of \( g = 10 \) nm.

It should also be noted that only spontaneous emission rate is speeded up by the concentrated slot mode, while the intrinsic nonradiative decay path related to the initial FQY \( \eta_0 \) keeps unchanged, leading to an enhanced FQY \( \eta_{\text{mod}} \). The discussion above has assumed a unit initial FQY (\( \eta_0 = 1 \)). For a finite FQY, however, we find that the spontaneous emission enhancement can actually enhance the FQY. In the slot waveguide, the modulated (enhanced) FQY \( \eta_{\text{mod}} \), describing the ratio of the radiation, can be calculated by

\[
\eta_{\text{mod}} = \frac{\eta_0(1 + \bar{F}_p/n_1)}{(1 - \eta_0) + \eta_0(1 + \bar{F}_p/n_1)}. \tag{10}
\]

As demonstrated in Figure 4(c), for an initial \( \eta_0 = 0.5 \), the modified FQY \( \eta_{\text{mod}} \) exceeds 0.8 over a broad wavelength range in the case of \( g = 10 \) nm.

We turn to analyze the total photon conversion efficiency of this LSC. Besides the isotropically averaged waveguide coupling efficiency \( \bar{\beta} \) and the modified FQY \( \eta_{\text{mod}} \), the total efficiency \( \eta_{\text{LSC}} \) should also include the solar photon absorption efficiency \( \eta_{\text{abs}} \), the transportation efficiency \( \eta_{\text{tran}} \) from emitters to the PV systems. Thus, we have

\[
\eta_{\text{LSC}} = \eta_{\text{abs}}\eta_{\text{mod}}\bar{\beta}\eta_{\text{tran}}. \tag{11}
\]

In the following we will explain how the slot waveguides play significant roles in obtaining a high \( \eta_{\text{LSC}} \). First, \( \eta_{\text{abs}} \) depends on the intrinsic property of the active medium, such as the absorption cross-section and the absorption spectrum. Interestingly, the absorption ability of single emitters is expected to be improved due to the enhanced emission with a high \( \bar{F}_p \) in the slot waveguide. A higher \( \bar{F}_p \) corresponds to a faster spontaneous emission of the active medium molecules, which implies the stimulated molecules can go back to their ground state faster and the round time of the absorption-emission process is significantly shortened \([21]\). In other words, the absorption length of the slot region is efficiently increased. Second, in Figure 4, both \( \eta_{\text{mod}} \) and \( \bar{\beta} \) are improved over a broad wavelength range, which can be further improved by aligning the dipoles of luminescent centers parallel to the electric field of quasi-TE modes \([22]\, [23]\). Third, the luminescent photons
should be transported to the LSCs edges where the PV systems can absorb these photons and convert them to photoelectrons. In this process, the photon transportation efficiency $\eta_{tran}$ can be high because the slot provides good propagating modes along z direction. Nevertheless, $\eta_{tran}$ may be degraded by the re-absorption phenomenon of the active medium. Potentially, the re-absorption can be suppressed by choosing luminescent centers whose absorption and emission spectra have a small overlap due to a large Stokes shift [24], or using luminesphors with unitary FQY which effectively close the nonradiative decay channel.

![Diagram](image)

**FIG. 5:** Schematic illustration of a tandem slot waveguide LSC. From top to bottom, the slot waveguides absorb solar photons with increasing wavelengths

Finally, it should be noticed that slot waveguides possess an optimized geometrical size for a specific wavelength band. For instance, if the thickness $a$ of high-index layers is too large, more energy will distribute in the high-index layers and less in the slot waveguide, so the Purcell effect of this structure will be dramatically weakened. As an example, if we increase the size $a \times b$ to $3a \times 3b$, for 550 nm wavelength in Figure 3(a), $F_\beta$ decreases from 11 to 1.4, and $\beta$ from 0.9 to 0.5. Nevertheless, an expanded geometry $a \times b$ is required to support propagating modes well in the slot waveguide for long wavelengths. To obtain the highest power efficiencies, on one hand, the absorption band can be broadened by mixing dyes and quantum dots with different sizes. On the other hand, it is also potential to construct tandem LSCs which can absorb more solar photons in a broad band. Incident solar photons are first absorbed by a LSC employing a short-wavelength active medium, such as a specific dye. Photons with longer wavelengths are transmitted through the first LSC and then absorbed by the second LSC employing a long-wavelength active medium and an expanded geometry $a \times b$, as shown in Figure 5. To fabricate such a LSC structure, a nanoimprinting technique can be utilized which can reach large areas up to several inches wide [25].

In summary, we present a theoretical description of nanoscale LSCs where the conventional ray-optics model is no longer applicable. Based on the Fermi-golden rule, we evaluate the spontaneous emission enhancement $F_\beta$ and the waveguide coupling efficiency $\beta$ in a slot waveguide consisting of a nanometer-sized low-index slot region sandwiched by two high-index regions. It is found that the slot waveguide provides a broadband enhancement of light harvesting by the luminescence centers in the slot region. In spite of a low initial FQY $\eta_0 = 0.5$, approximately 80% solar photons can re-emit as luminescent photons, and more than 80% luminescence can be coupled into the waveguide modes for ultimate absorption by the solar cell located on the waveguide edge. This LSC scheme holds a great potential to construct a tandem structure which can absorb nearly full-spectrum solar photons, and is of special interest for building integrated nano-solar-cell applications.

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