Nanowire Solar Cell Above the Radiative Limit

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A lossless solar cell operating at the Shockley–Queisser limit generates an open circuit voltage ($V_{oc}$) equal to the radiative limit. At $V_{oc}$, the highly directional beam of photons from the sun is absorbed and subsequently externally re-emitted into a $4\pi$ solid angle, providing a large photon entropy loss. A solar cell can beat the Shockley–Queisser limit and approach the 46.7% ultimate limit by decreasing the output solid angle of the light emission at open circuit conditions. Here, a design for an indium phosphide single nanowire solar cell capable to operate 159 mV above the radiative limit is presented. The spontaneous emission factor is first optimized into a guided mode of the nanowire toward 68%. The authors subsequently launch a guided mode at the bottom straight part of the tapered nanowire yielding a photon escape probability of 81% for a tapering angle of $\theta = 1.2^\circ$ and a top facet with a radius of 83 nm. When assuming homogeneous light emission along the nanowire, an outcoupling efficiency of 42% of the emitted light is obtained. The final optimization is the reduction of the emission cone toward $11 \times 10^{-3} \text{ sr}$ by focusing the guided mode with an external lens.

1. Introduction

A lossless solar cell with ideal contacts will operate at the Shockley–Queisser (S–Q) limit with a maximum photovoltaic conversion efficiency of 33.7% at a bandgap of 1.34 eV. At short circuit conditions, such a cell absorbs all sunlight below the material's bandgap and converts all absorbed photons into current. At open circuit conditions, absorbed photons do not contribute to the current and therefore have only two possibilities: either to be re-emitted externally or to recombine nonradiatively and therefore to heat up the cell. For an ideal solar cell without losses, external re-emission of all absorbed photons is the only option.

Even at the S–Q limit, a conventional solar cell is converting a highly directional beam of photons originating from the sun into a randomized isotropic emission with a $4\pi$ solid angle pattern. It is important to understand that the conversion of a parallel beam of photons into a random directional emission pattern constitutes a major thermodynamic loss due to a strong increase of the entropy of the photons. In a conventional solar cell, the increase of the photon entropy accounts for a large voltage loss of ≈0.3 V for a cell that is already operating at the S–Q limit. This voltage loss can be circumvented if all emitted photons at $V = V_{oc}$ are emitted into a narrow emission cone with a solid angle of $6.8 \times 10^{-5} \text{ sr}$, which is equal to the solid angle of the solar emission cone reaching the earth. In this case, the entropy of the photons is not increasing, thus eliminating a major thermodynamic loss mechanism.

A planar solar cell of, for example, GaAs is struggling with two problems which prevents the cell to reach or exceed the S–Q limit. The first problem is the small photon escape probability due to total internal reflection of photons at the front surface of the cell which is shown in Figure 1a by light gray cone. Photon escape is only allowed within a small internal solid angle, resulting in a <4% escape probability for each emission event within a planar layer of GaAs. For a solar cell featuring a very high internal radiative efficiency (IRE), photon recycling will allow multiple emission attempts, thus increasing the external radiative efficiency in excess of the 4% probability which applies for a single emission attempt. The second problem arises when looking to the emitted light outside the planar solar cell. Any absorbed photon will externally be re-emitted into a random direction making it impossible to sufficiently restrict the emission cone in such a way that all emitted radiation can, in principle, be focused back to the sun. In theory, the latter issue can be partly eliminated by limiting light emission angles using, for example, an angle restriction filter (Figure 1b). In practice, angle restriction increases the number of required photon recycling events, thus requiring an extremely large IRE inside the cell.

The photonic properties of a nanowire solar cell are fundamentally different from a planar solar cell since most of the light is emitted into guided modes. For a properly tapered nanowire, light moving in a guided mode from the bottom of the nanowire toward the small top diameter will gradually become less and less confined within the nanowire and more embedded into the surrounding air. This mechanism is referred to an adiabatic expansion of the guided fundamental HE$_{11}$ mode into the surrounding medium and is represented in Figure 1d–f. According to Figure 1d the electric field of the fundamental guided mode becomes less concentrated in the nanowire as the radius decreases. Effective refractive index,
reflection, and transmission calculations in Figure 1e,f show a decrease in refractive index and a decrease in reflection as the nanowire radius decreases as well as an increase in transmission with decreasing nanowire radius and therefore an efficient extraction of the mode into air. Friedler et al. calculated that the specular reflection at the nanowire top facet can be as low as 0.13%, while the back reflection due to tapering is only 0.6%.[9] Adiabatic expansion of the guided mode is expected to considerably enhance the external radiative efficiency in a nanowire solar cell, even for a single emission attempt.

An even more fundamental advantage of a nanowire solar cell is provided by the fact that a guided mode can be focused by using a lens. The light emitted by a nanowire solar cell can thus be emitted with a strongly reduced emission cone, by employing an external lens. The amount of light emitted into a guided mode is given by the spontaneous emission \( \beta \)-factor,[9] which will be shown to approach 70% in a properly designed nanowire. Our paper is distinctively different from previous papers attempting to reduce the emission cone of the emitted light. In these papers, photons are first emitted into random directions within the solar cell, thereby increasing photon entropy. An “external apparatus” is subsequently employed which only transmits photons transmitted into the correct direction.[7,8] Photons that are originally not emitted in the correct direction might be recycled to be eventually re-emitted in the correct direction. Our nanophotonic approach is not based on photon recycling. Instead, we are directly modifying the spontaneous emission directivity of the photons itself by using the fact that the spontaneous emission is either inhibited or enhanced by the presence of photonic modes in its vicinity.[10,11] A semiconductor nanowire is naturally guiding the photonic mode, allowing to enhance the directional emission probability since photons are preferentially emitted, with a 70% spontaneous emission factor, into a single guided nanowire mode.

The emission of light into a guided mode is a key to prevent an increase in photon entropy. In an ideal nanowire solar cell, the solar light is first absorbed while propagating in a guided mode and will subsequently be re-emitted into the same guided mode, in principle allowing to focus all light back to the sun, without increasing light entropy. In this paper, we quantitatively investigate the potential of a single nanowire solar cell for beating the S–Q limit, assuming perfect material properties with an IRE or photoluminescence quantum yield of 100%. Such a high IRE is not unrealistic since multiple authors have reported IREs above 99.5% for GaAs[12,13] and of 97% for indium phosphide (InP).[14]

It should be noted that the solar light absorption properties of a nanowire solar cell have already been investigated.[15–18] The light absorption in a 2 \( \mu \text{m} \) length tapered InP nanowire has been calculated to be as high as 98% of the incoming solar spectrum with a photon energy above the materials bandgap. The light absorption in a nanowire is actually larger than in a planar solar cell due to a reduced light reflection at the top surface of the nanowire and enhanced light-trapping through the resonant coupling to leaky waveguide modes.[19–21] A large light absorption was experimentally demonstrated in an InP nanowire solar cell with an ITO top contact, featuring an average light absorption efficiency of 90%.[22] This InP nanowire solar cell features a power conversion efficiency of 17.8% which was mainly limited by a \( V_{oc} \) of 272 mV below the radiative limit. The short circuit current of the nanowire solar cell was 29.3 mA cm\(^{-2}\) while the record planar InP solar cell features a short circuit current of 30.5 mA cm\(^{-2}\), thus showing that an optimization of the open circuit voltage deserves the highest priority.

2. Spontaneous Emission Factor

We first calculate the dispersion relations of the first three fundamental modes (TE\(_{01}\), TM\(_{01}\), and HE\(_{11}\)) in infinitely long InP nanowires with finite difference time domain simulations (Figure S1, Supporting Information) for 920 nm wavelength.
as shown in Figure 2a. Above the light line, which is represented by a horizontal dashed black line, all the modes are guided; while below the light line, they are leaky implying that the mode has an effective refractive index below the refractive index of surrounding medium (air in our case). Leaky modes radiate into the surrounding medium whereas guided modes with effective refractive index higher than 1 are guided inside the nanowire. From analytical calculation of the emission from an infinite nanowire assuming homogeneously distributed spontaneous currents, 100% light extraction efficiency can be obtained when the radius of the nanowire is small enough to allow only a single guided mode.[23] We are thus mostly interested in the fundamental HE$_{11}$ mode. For too small diameter, this mode becomes however weakly guided when it approaches the light line, implying that we should use a nanowire radius above $\approx$100 nm.[24] To maximize the photon escape probability it is required to efficiently extract the guided mode from the nanowire into the air. It has been shown that a guided mode can be adiabatically expanded into the surrounding air with very low loss by using a tapered nanowire with small tapering angle.[16,25] Moreover, to restrict the emission cone of the emitted light, the polarization of the dipole depends on the selection rules for the dipole transition, resulting in a random polarization for a zincblende structure.[26] It is known that it is possible to obtain a $\beta$-factor very close to unity for horizontally oriented dipole emitters located exactly on the nanowire axis.[9]

The $\beta$-factor for an infinitely long InP nanowire as a function of the nanowire radius is calculated in Figure 2b for different radial positions of the dipole averaged over three perpendicular orientations, and using an emission wavelength of 920 nm close to the InP bandgap. The black curve corresponds to an on-axis dipole and the orange curve corresponds to a dipole located on the nanowire sidewall.

When the dipole moves off-center, the $\beta$-factor first decreases toward 0.6 and subsequently increases again, which is due to an interplay of guided modes for perpendicularly and parallel oriented dipoles at different positions along the nanowire radius. An on-axis dipole oriented perpendicular to the nanowire axis couples to the HE$_{11}$ mode, whereas a dipole oriented parallel to the axis couples to the TM$_{01}$ mode. The $\beta$-factor is calculated as...
a function of both the nanowire radius and the relative radial dipole position, which is shown in Figure 2c. The coupling to the HE11 mode is the highest (>87% of emitted light) for a nanowire radius between 95 and 140 nm with a dipole positioned close to the center of the nanowire. For a radius between 117 and 133 nm, more than 75% of the light emitted near the sidewall of the nanowire couples to the fundamental mode with a maximum for a radius of 125 nm (Figure 2b,c). When we average over all dipole positions and orientations for a nanowire with a radius of 125 nm, the β-factor is found to be 68%, implying that this fraction of the emitted light is coupled to the fundamental mode which can in principle be redirected back to the sun.

3. Photon Escape Probability

We subsequently perform a nanophotonic geometry optimization of the photon escape probability $P_{\text{esc}}$ which is related to the open circuit voltage by $V_{\text{oc}} = V_{\text{oc}}^\text{rad} - \frac{kT}{q} \ln \left( \frac{P_{\text{esc}}}{P_{\text{net}}} \right)$. We use a single nanowire with a bottom straight section with a height in the range of 0.2–1.0 μm and a top tapered section with a height in the range of 1–2 μm (Figure 3a) keeping the total nanowire length fixed at 2 μm. We include a silver bottom mirror to redirect the emitted light toward the top facet. The length of the nanowire is chosen to be long enough to absorb all incident solar radiation by an array of the nanowires (Figure S7, Supporting Information) and adiabatically expand the fundamental mode into air, while keeping the length smaller than the minority carrier diffusion length to achieve an efficient current generation. The tapered part is chosen long enough to maintain the adiabaticity for the guided mode. For optimizing the tapering, the dipole emitter is initially positioned in the center of the bottom section. The calculations will be later extended to an average over radial and axial dipole positions. Tapering minimizes the top facet reflection at the small diameter nanowire tip and maximizes light absorption using a large bottom diameter. The main role of the tapered section at the top is to adiabatically expand the mode into air, which improves the light outcoupling, besides reducing the reflection. Adiabatic expansion of the mode requires a very slow variation in the effective index of the guided mode, which is achieved by a very slow variation of the nanowire radius.

The photon escape probability through the top facet of the nanowire is given in Figure 3b as a function of the height of the bottom part of the nanowire and the tapering angle, and is defined as the ratio between the emitted power from the top facet to the total emitted power of the dipole inside the nanowire as a function of the bottom radius and the tapering angle $\theta$. The photon escape probability as a function of the bottom radius showing saturation at 120 nm.

![Figure 3](image-url)

**Figure 3.** a) Nanowire geometry with a bottom straight part and top tapered part. A horizontally oriented dipole is positioned in the center of the bottom straight part. b) Photon escape probability from the top facet of the nanowire as a function of the height of the bottom straight part and the tapering angle $\theta$ for a bottom radius of 125 nm. c) Photon escape probability from the top facet of the nanowire as a function of the height of bottom part for a fixed tapering angle $\theta = 1.2^\circ$ (black curve) and photon escape probability as a function of tapering angle for a fixed height of bottom part of 200 nm (blue curve). d) Photon escape probability as a function of the bottom radius and the tapering angle $\theta$. e) Photon escape probability from the top facet of the nanowire as a function of the tapering angle for a fixed bottom radius of 120 nm (black curve) and photon escape probability as a function of the bottom radius for a fixed tapering angle $\theta = 1.2^\circ$ (blue curve). f) Photon escape probability as a function of the bottom radius showing saturation at 120 nm.
nanowire. We obtain the highest photon escape probability for a small range of tapering angles between 0.8° and 1.5° and a short height of the bottom part (≤0.3 µm). The photon escape probability as a function of tapering angle for a height of the bottom part of 0.2 µm and the photon escape probability as a function of the height of the bottom part for a tapering angle of 1.2° are shown in Figure 3c as blue and black lines, correspondingly. A maximum photon escape probability is observed for a tapering angle of 1.2° for a chosen height of the bottom part of 0.2 µm. For a height of the tapered top (bottom) section of 1.8 µm (0.2 µm), the photon escape probability from the top facet is calculated in Figure 3d as a function of the bottom radius of the nanowire and the tapering angle. For small tapering angles, additional features occur because the top diameter becomes of the order or even smaller than the mesh step. The maximum photon escape probability is obtained for a bottom radius above 95 nm and tapering angles ranging from 0.5° to 2°, which corresponds to the range of nanowire diameters yielding the highest β-factor (Figure 2). Figure 3e shows the photon escape probability as a function of the bottom radius for a tapering angle of 1.2° showing a maximum for a radius of 120 nm (blue curve) as well as the photon escape probability as a function of the tapering angle (black curve) for a bottom radius of 120 nm with a maximum for tapering angle of 1.2°. The photon escape probability as a function of the bottom facet radius is shown in Figure 3f, illustrating the saturation at a radius above 120 nm. By choosing the radius of the bottom facet equal to 120 nm, we find a maximum photon escape probability of 81% for a tapering angle of θ = 1.2°, which corresponds to the top facet radius of 83 nm.

We now calculate the averaged photon escape probability at open circuit for a tapered nanowire with the parameters established above. We assume homogeneous absorption across the nanowire length (Figure S8, Supporting Information). We now average over horizontal and vertical positions and orientations of the dipole emitter within the tapered nanowire. In this case 42% of light will be emitted from the top facet and can be collected with the top lens. Here, we do not take into account emission from the sidewalls because this light will not be collected with the top lens. We emphasize that this number is actually a lower limit ignoring photon recycling. If we calculate Pesc from the whole structure except the bottom facet, 96% of light will be emitted from a nanowire standing on a silver mirror. Part of the emitted light from a side facet of a neighboring nanowire might thus be absorbed and recycled in the nanowire under study, thus increasing the photon escape probability. For a tapered nanowire standing on a native InP substrate, we calculate a photon escape probability from the top facet of 21%, while Pesc from the whole structure except bottom facet is 53%. The latter result was calculated for the case of a lossy nanowire with a nonzero imaginary part of the refractive index which is the most realistic case. Because of the nonzero imaginary part of refractive index for the bandgap wavelength of 920 nm, the emitted light will be partially absorbed by the structure. The photon escape probability of 53% is exactly the same as for straight nanowires but higher than for nanocones.[28] However, in the work of Anttu et al., the imaginary part of the refractive index was assumed to be zero.

4. Reduction of the Emission Cone

The second step in maximizing the Voc is by limiting the emission cone of the outcoupled light. The Voc can be expressed as

\[ V_{oc} = V_{oc}^{\text{Ultimate}} \left( \frac{kT}{q} \ln \frac{\varepsilon_{\text{in}}}{\varepsilon_{\text{out}}} \right) \left( \frac{kT}{q} \ln(\eta_{oc}^*) \right), \]

in which \( \varepsilon_{\text{in}} \) is the input étendue of the sun, \( \varepsilon_{\text{out}} \) is the étendue of the solar cell, \( \eta_{oc}^* \) is the average over horizontal and vertical positions and orientations of the dipole emitter within the tapered nanowire. In this case 42% of light will be emitted from the top facet and can be collected with the top lens. Here, we do not take into account emission from the sidewalls because this light will not be collected with the top lens. We emphasize that this number is actually a lower limit ignoring photon recycling. If we calculate Pesc from the whole structure except the bottom facet, 96% of light will be emitted from a nanowire standing on a silver mirror. Part of the emitted light from a side facet of a neighboring nanowire might thus be absorbed and recycled in the nanowire under study, thus increasing the photon escape probability. For a tapered nanowire standing on a native InP substrate, we calculate a photon escape probability from the top facet of 21%, while Pesc from the whole structure except bottom facet is 53%. The latter result was calculated for the case of a lossy nanowire with a nonzero imaginary part of the refractive index which is the most realistic case. Because of the nonzero imaginary part of refractive index for the bandgap wavelength of 920 nm, the emitted light will be partially absorbed by the structure. The photon escape probability of 53% is exactly the same as for straight nanowires but higher than for nanocones.[28] However, in the work of Anttu et al., the imaginary part of the refractive index was assumed to be zero.
the external radiative efficiency, and $V_{\text{ultimate}}$ is the ultimate open circuit voltage which is slightly larger than the semiconductor bandgap.[16] The étendue is defined as $\varepsilon = A\Omega$, where $A$ is the area of the emitter and $\Omega$ is the solid angle of the emission. An external lens is expected to collimate the emitted light or to match $\varepsilon_{\text{out}}$[29] and the input étendue of the solar light, $\varepsilon_{\text{in}}$, which is explained in detail in Figure S4, Supporting Information. For this purpose a simple dielectric plano–convex microlens design with a refractive index $n = 1.5$ is investigated. These lenses have already been fabricated using a resist reflow process.[30] The working principle of such a lens is based on the so called photonic nanojet effect which is a high-intensity and spatially localized region due to the light scattering on the dielectric spherical particles with dimensions higher than the wavelength of light. A photonic nanojet is well described by Mie theory, which shows that the focal point for a sphere with refractive index higher than 2 is located inside the sphere, whereas the focus is located on the boundary of the sphere for a refractive index between 1 and 2.[31] However, if we consider a truncated sphere instead of a sphere, we can shift the focal point further away from the boundary of the sphere, depending on the cutting height of the truncated sphere.[13] We investigate a plano–convex truncated sphere lens to reduce the emission angle of a nanowire solar cell (Figure 4a).

We calculate the open circuit voltage of the nanowire solar cell using optimized photon escape probability and reduced directivity obtained by using a truncated spherical lens. We assume an IRE of 100% and use a photon escape probability of 42% as calculated above. When using a lens with 4.6 $\mu$m diameter (Figure 4b), the emission angle of the nanowire emission is decreased from $840 \times 10^{-3}$ to about $24 \times 10^{-3}$ sr (Figure 4c,d), which yields an open circuit voltage of 1176 mV. This open circuit voltage is 139 mV higher than the radiative limit but still 174 mV less than the ultimate limit. For a lens diameter of 8 $\mu$m, the emission angle is further decreased to $11 \times 10^{-3}$ sr, resulting in an open circuit voltage of 1196 mV, which is even 159 mV higher than the radiative limit and only 154 mV below the ultimate limit. The same calculations can be done for a nanowire standing on a native substrate. Photon escape probability from the top facet for this case is equal to 21% and the open circuit voltage equals to 1158 mV for a 4.6 $\mu$m diameter lens which is 121 mV higher than radiative limit and 192 mV lower than ultimate one. For an 8 $\mu$m lens, the open circuit voltage is equal to 1178 mV which is 141 mV higher than the radiative limit and 172 mV lower than the ultimate limit.

5. Conclusions

In conclusion, the light management in a single InP nanowire photovoltaic cell is optimized for increasing the open circuit voltage above the radiative limit. In a straight nanowire with a radius of 125 nm, 68% of the emitted light can be coupled to the fundamental guided mode. Light emission from the guided mode is essential to be able to efficiently collimate the emitted light to the sun. We subsequently performed a nano-photonic optimization of the light emission from the top facet of the nanowire. For an optimal nanowire tapering, 42% of the light can be emitted from the top facet, thus approaching the radiative limit. We finally studied the collimation of the guided light with a plano–convex microlens. The emission angle can be decreased from $840 \times 10^{-3}$ to $11 \times 10^{-3}$ sr using a 8 $\mu$m diameter lens, which allows beating the radiative limit with 159 mV and approach the ultimate limit voltage.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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