RESEARCH ARTICLE

Limiting efficiencies for intermediate band solar cells with partial absorptivity: the case for a quantum ratchet

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ABSTRACT

The intermediate band solar cell (IBSC) concept aims to improve upon the Shockley–Queisser limit for single bandgap solar cells by also making use of below bandgap photons through sequential absorption processes via an intermediate band (IB). Current proposals for IBSCs suffer from low absorptivity values for transitions into and out of the IB. We therefore devise and evaluate a general, implementation-independent thermodynamic model for an absorptivity-constrained limiting efficiency of an IBSC to study the impact of absorptivity limitations on IBSCs. We find that, due to radiative recombination via the IB, conventional IBSCs cannot surpass the Shockley–Queisser limit at an illumination of one Sun unless the absorptivity from the valence band to the IB and the IB to the conduction band exceeds \( \approx 36\% \). In contrast, the introduction of a quantum ratchet into the IBSC to suppress radiative recombination can enhance the efficiency of an IBSC beyond the Shockley–Queisser limit for any value of the IB absorptivity. Thus, the quantum ratchet could be the vital next step to engineer IBSCs that are more efficient than conventional single-gap solar cells. © 2016 The Authors. Progress in Photovoltaics: Research and Applications published by John Wiley & Sons, Ltd.

KEYWORDS

intermediate band solar cell; absorptivity; limiting efficiency; quantum ratchet

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1. INTRODUCTION

The intermediate band solar cell (IBSC) (Figure 1(a)) is a high-efficiency solar cell concept with a detailed balance efficiency limit of 63% [1] at full concentration, higher than the Shockley–Queisser limit of 41% [2] for conventional single-bandgap solar cells. The main implementations of IBSCs so far have been based on semiconductor nanostructures such as quantum dots (QDs) [3], bulk semiconductor highly mismatched alloys [4], and bulk semiconductor materials containing a high density of deep-level impurities [5]. Using these approaches, the key IBSC operating principles have been demonstrated [6,7]; however, the Shockley–Queisser limit is still far from being exceeded in practice. Although IBSCs have been reported with efficiencies marginally higher than that of an equivalent single-gap reference device [8–10], in most cases, reported efficiencies are actually lower than the equivalent single-gap reference cell [11–16]. This is because, in present implementations, the improvement in photocurrent offered by the intermediate band (IB) absorption is accompanied by a much larger reduction in open circuit voltage caused by increased recombination via the IB levels.

To alleviate the problem of additional recombination, the concept of a quantum ratchet IBSC (QR-IBSC) was recently introduced [17] (Figure 1(b)). Analogous to a mechanical ratchet, the QR consists of an efficient and thermodynamically irreversible scattering step that takes electrons from the states in the IB (which are optically connected to the valence band (VB)) to states in the ratchet band (RB) that are optically connected only to the conduction band (CB). Optical transitions from the RB to the VB are considered to be forbidden by symmetry and/or spatial separation of electronic wavefunctions. An example of a naturally occurring QR is dyes used for molecular upconversion solar cells [18], which have recently also been employed directly in IBSCs [19]. Even though the ratchet step introduces an energy loss, such a QR...
configuration drastically reduces radiative recombination and can thereby increase the efficiency of an IBSC in the radiative limit [17].

The radiative limit for IBSCs [1] contains the assumption that the absorptivity, defined as the fraction of incoming light that is absorbed by the device, from the VB to the IB and from the IB to the CB is both unity in their respective energy ranges. However, many implementations of the IB concept that have been proposed operate far from this ideal configuration. Usually, the IB absorptivity is less than 1%, meaning that only a very small amount of the energy in the relevant spectral range is captured by the electronic system. Some calculations on the impact of low absorptivity have been made for specific material systems or bandgap combinations [20–23], and it has been recognized that, in those cases, high solar concentrations, high absorptivities, and possibly light trapping are needed to derive any real efficiency benefit from the IB.

Here, we systematically investigate the impact of the limited absorptivity of the IB on the efficiency of IBSCs by defining and evaluating an absorptivity-constrained limiting efficiency (ACLE), where the absorptivity of transitions into and out of the IBSC is limited to a certain value $a_{VB} = a_{VC} = a_{IB} \leq 1$, while the absorptivity of the VB to CB transition is set to unity, that is, $a_{VC} = 1$. Hence, this ACLE models the IBSC as a blackbody at energies above the VB–CB bandgap, as a greybody, with absorptivity $a_{IB}$, at energies from the IB to CB separation up to the VB to CB separation and as transparent to light at lower energies (see Figure 1(c) for an illustration of this concept), while it considers an ideal, radiatively limited device in all other aspects. Note that we have assumed, without loss of generality, that the IB is closer in energy to the CB than to the VB, that is, $E_{CB} - E_{IB} < E_{CB} - E_{VB}$. We find that at one Sun, and for $a_{IB} < 0.36$, any standard IBSC bandgap configuration (Figure 1(a)) results in an ACLE below the Shockley–Queisser limit. We compare this to the ACLE of the QR-IBSC concept, and we show that the QR-IBSC can overcome the Shockley–Queisser limit even for low concentrations and low values of $a_{IB}$. For concentrations below $C \approx 23000$, the QR-IBSC is more efficient than an equivalent conventional IBSC for all values of the absorptivity $a_{IB}$.

Section 2 explains the model used to achieve the results presented in Section 3. Conclusions from this study are drawn in Section 4.

## 2. Model

In our model, we make the usual radiative limit assumptions for the IBSC, that is, that non-radiative recombination can be neglected and that the absorption of light takes place via three different transitions, none of which overlap spectrally: the transition from the VB to the CB, the transition from the VB to the IB, and the transition from the IB or RB (for the QR-IBSC) to the CB. For the latter two transitions, involving the IB and RB, we set the absorptivities, $a_{IB}$, to be equal and study the effects of varying them.
For the efficiency calculations, we define carrier generation rates [17,24] $G_{VB}$ for VB to IB transitions

$$G_{VB} = \frac{2f a_{IB}}{h^2 c^2} B_2(0, T_{sun}, E_{IB} - E_{VB}) + \frac{2(\pi - f) a_{IB}}{h^2 c^2} B_2(0, T_c, E_{CB} - E_{IB} + dE, E_{IB})$$  \hspace{1cm} (1)

$G_{IC}$ for IB (RB) to CB transitions

$$G_{IC} = \frac{2f a_{IB}}{h^2 c^2} B_2(0, T_{sun}, E_{CB} - E_{IB} + dE, E_{IB}) + \frac{2(\pi - f) a_{IB}}{h^2 c^2} B_2(0, T_c, E_{CB} - E_{IB} + dE, E_{IB})$$  \hspace{1cm} (2)

and $G_{VC}$ for valence to CB transitions

$$G_{VC} = \frac{2f a_{IB}}{h^2 c^2} B_2(0, T_{sun}, E_{CB}, \infty) + \frac{2(\pi - f) a_{IB}}{h^2 c^2} B_2(0, T_c, E_{CB}, \infty)$$  \hspace{1cm} (3)

with the Bose integral of second order

$$B_2(\mu, T, E_0, E_1) = \int_{E_0}^{E_1} \frac{E^2}{E_{kB} T} \exp \left( \frac{E_{kB} T}{E} - 1 \right)$$  \hspace{1cm} (4)

The same absorptivity $a_{IB}$ affects the radiative recombination rates

$$R_{VB} = \frac{2\pi a_{IB}}{h^2 c^2} B_2(\mu_{VB}, T_c, E_{CB})$$  \hspace{1cm} (5)

$$R_{VC} = \frac{2\pi a_{IB}}{h^2 c^2} B_2(\mu_{CB}, T_c, E_{CB})$$  \hspace{1cm} (6)

$$R_{IC} = \frac{2\pi a_{IB}}{h^2 c^2} B_2(\mu_{CB}, T_c, E_{CB} - E_{IB} + dE, E_{IB})$$  \hspace{1cm} (7)

where the chemical potential differences between the bands, $\mu_{ij}$, determine the strength of the radiative recombination. We assume that the RB and IB possess a common quasi-fermi level, achieved through efficient scattering between the two sets of states.

The resulting rate equations, which describe the characteristics of the device, are given by

$$G_{VB} - R_{VB} = G_{IC} - R_{IC}$$  \hspace{1cm} (8)

$$qV = \mu_{VC} = \mu_{VB} + \mu_{IC}$$  \hspace{1cm} (9)

$$J = q(G_{VC} - R_{VC} + G_{IC} - R_{IC})$$  \hspace{1cm} (10)

with the voltage $V$ and the current density $J$. Equation (8) is solved numerically while imposing the condition given by Equation (9), to determine the quasi-equilibrium division between the chemical potentials of the cell, for a given applied voltage and concentration factor $C$. The result can be used to determine the current density $J$. The maximum of the product of current density $J$ and voltage $V$ divided by the solar Flux $U_{sun}$

$$U_{sun} = \frac{2f}{h^2 c^2} \int_0^{\infty} dE \exp \left( \frac{E^3}{E_{yb} T_{sun}} - 1 \right)$$  \hspace{1cm} (11)

then gives the power conversion efficiency of the solar cell at the operating point. Note that in this model, we have constrained the IB offset from the CB to be at least $6kT_{cell}$ to avoid significant thermal exchange between those two bands. If the IB gets too close to the CB, and thermal exchange becomes important, the operation of the device becomes indistinguishable from a single gap cell.

The difference between the equations for the situation containing a QR and without the QR lies only in the spectral portion of the light that is considered to be absorbed and drives the IB to CB transition and in which radiative recombination is going to occur. In the case of the QR-IBSC, the sum of the energies of the two photons driving the sequential absorption has to be at least $E_{CB} + dE$ instead of just $E_{CB}$, and this results in an energy loss. Because, in the radiative limit, the ratchet is described purely by the integration limits on generation and recombination rates, the same limiting efficiency calculations and all the results discussed in the succeeding texts also apply to a system in which the energy relaxation step happens in the CB or in the VB. We have illustrated the possible ratchet configurations in Figure 2, but, for definiteness, our discussions will focus on the configuration with the QR in the IB.

A QR that follows the aforementioned mathematical model has to fulfill three conditions. Firstly, there needs to be an energy relaxation step, $dE$, between IB and RB. Secondly, scattering between IB and RB needs to be faster than generation and recombination rates involving the IB, so that a common quasi-fermi level for the two subbands can be established, and thirdly, radiative transitions from RB to VB and CB to IB need to be forbidden. This can be achieved either via vanishing spatial overlap of wavefunctions or through optical selection rules stemming from the symmetry of the wavefunctions.

**Figure 2.** A schematic of the possible ratchet configurations with allowed transitions indicated as arrows. (a) A quantum ratchet (QR) in the intermediate band (IB), (b) a QR in the conduction band (CB), and (c) a QR in the valence band (VB), where the energy step now points upwards because holes are being scattered from VB to ratchet band (RB).
The CB ratchet configuration could be achieved by surrounding the QDs in a QD-IBSC with an energy barrier, thus spatially decoupling the QDs from the lowest energy CB states. A similar system, with the barrier extending across the plane of the device, has been investigated in [25]. However, one has to take care not to reduce electron mobility in the CB too much, while mobility in the RB and IB is not strictly necessary for the functionality of the device. Another possibility of implementing an energy relaxation step in the CB could be provided through spin-orbit coupling in dilute magnetic semiconductors as has been suggested by Olsson et al. in [26].

3. RESULTS

Figure 3 shows the ACLE plotted against the absorptivity $a_{IB}$ of transitions into and out of the IB (Figure 1(c)) for different types of IBSCs operating at one Sun. For each absorptivity value, $a_{IB}$, we calculated the efficiencies at the operating voltage of the ideal devices as a function of the energetic position of the different bands in steps of 0.01 eV. We then obtained the limiting efficiency for a given $a_{IB}$ as the maximum of these efficiencies. Here, one has to constrain $dE \leq 2E_{IB} - E_{CB}$, so that the RB to CB transition does not overlap spectrally with the VB to IB transition. As $a_{IB}$ is increased from 0, the ACLE for the conventional IBSC (red-solid line) initially decreases from the single gap limit; it only recovers the single gap value at an absorptivity $a_{IB}$ of around 36% and eventually reaches a value of 46.3%, far above the single gap limit.

From the low $a_{IB}$ behavior of the ACLE, we can see how the introduction of a low-absorptivity IB, and thereby an additional radiative recombination channel, actually degrades the performance of a device. In a low-absorptivity IB, only relatively few electrons will make the two-step transition, while more electrons that have made the direct transition between CB and VB can relax via the two-step recombination route. This effect arises purely from basic thermodynamic principles and is independent of the specific implementation. An example of this behavior for a specific QD IB system can be found in [22]. Because our calculation does not include the effects of any device imperfections or of non-radiative recombination, it tells us that, unless $a_{IB} > 0.36$, a conventional IBSC operated
at one Sun can never be more efficient than an optimal single gap cell. Typically, IBSC devices operate at values of absorptivity $a_{IB}$ that are orders of magnitude lower than this.

In contrast, the QR-IBSC in the radiative limit (blue-dotted line in Figure 3) does not show any such degradation because of the insertion of a low-absorptivity IB. Its ACLE immediately rises from the single gap limit as $a_{IB}$ is increased from 0. The introduction of an IB in conjunction with a QR thus helps to improve the performance of the radiatively limited solar cell even for very low values of $a_{IB}$. This is because the ratchet step suppresses the build-up of thermal occupation in the higher state in the IB and thus suppresses the radiative recombination from the CB to the VB via the IB.

The result for the conventional IBSC (Figure 3) shows a kink at a value of $a_{IB}$ of around 24%. This kink occurs because the optimal cell configuration changes its mode of operation from essentially a single gap cell operation to a proper IBSC operation with sequential photon absorption in the IB becoming crucial for its efficiency. The crossover between these regimes manifests itself, in the optimal bandgaps shown in Figure 4(a), as a sudden and simultaneous increase in $E_{CB}$, a decrease in $E_{IB}$, and an increase in the operating voltage.

We see in Figure 4 that the ideal bandgap combination, for both a conventional IBSC and a QR-IBSC, strongly depends on the value of $a_{IB}$, that is, the more effectively the spectrum below the VB to CB bandgap is absorbed, the larger the optimal bandgap. For very small $a_{IB}$ value, the ideal VB to CB bandgap is very close to the ideal bandgap for a single gap cell, while the ideal VB to CB separation is much larger for high values of $a_{IB}$. We also see that, at first, a rise in $a_{IB}$ has the effect of lowering the operating voltage, until the kink is reached, at which point the IBSC begins to work properly, with a voltage that is above the IB to VB separation. As $a_{IB}$ is further increased, the subsequent rise in the ideal operating voltage is mainly a consequence of the increase in the VB to CB bandgap of the optimal device. For the QR-IBSC, the rise of optimal bandgaps with $a_{IB}$ occurs more smoothly. The optimal ratchet step, $dE$, decreases slightly because of the change in the differential pay-off between the suppression of radiative recombination and the electron energy loss introduced by the ratchet step. These results show how important it is to take realistically achievable $a_{IB}$ values into account when thinking about appropriate bandgaps for an IBSC.

In Figure 5(a) it is shown how the radiatively limited efficiency of an IBSC configuration, which is optimized for specific values of $a_{IB}$, changes with the $a_{IB}$ values that are achieved. In particular, the bandgap combination optimized for unity absorptivity, $a_{IB} = 1$, performs very badly if $a_{IB}$ is instead much lower; it drops below 20% for values of $a_{IB}$ close to 0 because of large below bandgap loss. On the other hand, the performance of a single bandgap cell, with a comparatively large bandgap, of 2.4 eV, can be improved by including absorptive IB states, even if they have only very low $a_{IB}$ values. Such an improvement (albeit from very low efficiency values) has been observed in [10] for a ZnTe:O cell.

The slope of the efficiency versus $a_{IB}$ curve is smaller for bandgap combinations that are optimized for smaller values of $a_{IB}$. This can easily be understood if one considers that the ideal separation between VB and CB increases with $a_{IB}$. For a single gap cell, this increase in bandgap means that, simultaneously, the below bandgap loss is increased, and the thermalization loss is decreased [27]. Because the IB concept helps to alleviate below bandgap loss, the differential gain obtained by increasing $a_{IB}$ improves for larger VB to CB separations. If the value of $a_{IB}$ is rather small, the below bandgap loss is much greater than in the ideal single gap cell, and the efficiency is reduced from the single gap limit, with radiative recombination compounding the problem. For small VB to CB separations, the IB cannot possibly deliver a strong benefit because the loss mechanism of the equivalent single gap cell is weighted more towards thermalization loss then towards below bandgap loss. Note the crossover in the curves in Figure 5(a), the design with the lower bandgap is the more efficient one at low $a_{IB}$ values, but this reverses for higher $a_{IB}$.

The operating voltage $V_{op}$ of the conventional IBSC, using the bandgap configuration optimized for $a_{IB} = 0.6$
as an example, is shown in Figure 5(b). Here, increasing $a_B$ decreases the operating voltage. This is in contrast to the result in Figure 4(a) where the operating voltage increases with absorptivity $a_B$. This is because the global optimization process in the ACLE calculation results in bandgaps in Figure 4 that increase with $a_B$, whereas they are kept constant in Figure 5(b). For fixed bandgap combinations, the observed decrease in operating voltage $V_{op}$ as $a_B$ increases is due to the increased radiative recombination; the efficiency of the cell still rises because the current increase more than compensates for the decrease in operating voltage.

Up to now, we have considered different IBSCs only under one Sun ($C = 1$) illumination. Often, one would want to employ IBSCs in concentrator solar cells. In this case, the role of radiative recombination is significantly reduced from the one Sun case. The ACLE for a concentration factor of $C = 500$, shown as solid line in Figure 6, rises immediately for very low values of $a_B$ even though the IB transitions then absorb only a small part of the relevant spectrum. The crossover from efficiency reduction for small values of $a_B$, compared with the single gap efficiency, to immediate efficiency increase, occurs at a concentration factor of $C \approx 30$. A QR-IBSC (blue-dotted line) can, however, still give a small advantage over the conventional IBSC configuration even at $C = 500$, albeit with smaller values of the energy step $dE$. For unity absorptivity, the advantage of any ratchet step disappears at a concentration of $C \approx 23000$, and at full concentration, the ratchet step results in an overall efficiency loss for the radiatively limited device.

![Figure 6](image_url). Limiting efficiencies versus absorptivity $a_B$ for intermediate band solar cells (IBSCs) at a concentration of $C = 500$. The dashed black line indicates the limiting efficiency of a single gap solar cell.

### 4. CONCLUSIONS

In summary, we have introduced and analyzed the concept of an ACLE of IBSCs. This concept enables us to clarify the important role of radiative recombination through the IB in the efficiency reduction that is observed in currently demonstrated IBSCs. Indeed, at one Sun illumination, the radiative limit efficiency for an IBSC with only small absorptivity $a_B$ from the VB to the IB and the IB to the CB is smaller than the radiative limit efficiency for a single gap solar cell.

There are two possible approaches to this problem. The first, which is one of the main topics of present IB research, is to increase the absorptivity $a_B$ towards unity. This requires a significant increase in the density of IB states beyond what is presently achieved in most implementations and can be assisted by optical absorption enhancement techniques. The second, more recently proposed, approach is the introduction of a QR mechanism. This ratchet eliminates radiative recombination from the CB to the VB via the IB, and here, we find that, in the radiative limit, it allows any small increase in photo-current because of an absorptivity in the IB to be translated directly into additional solar cell efficiency. Implementing both of these approaches together promises the highest efficiencies for IBSCs.

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