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Comment on “Analysis of Quantum Coherent Semiconductor Quantum Dot p-i-n Junction Photovoltaic Cells”

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According to conventional wisdom, radiative recombination limits the quantum efficiency of a photocell; e.g., when illuminated by a monochromatic beam of solar photons (characterized by frequency ν_s and temperature T_s) this limit is $eV/\hbar\nu_s = \eta_c$ where V is the induced voltage, $\eta_c = 1 - T_a/T_s$ and T_a is the ambient temperature. As stated in a recent review [1]:

> "That leaves radiative recombination as the major [energy loss] process. Can this be avoided? The answer is no. If a radiative upward transition to generate the excitation is allowed, its reversal, the radiative downward transition must be allowed as well."

However in [2], it is shown that it is possible to break detailed balance as in the case of lasing without inversion in quantum optics [3] and the photo-Carnot engine in quantum thermodynamics; yielding a photocell quantum efficiency $eV/\hbar\nu_s = \eta_c + \delta\eta$ where $\delta\eta$ represents a model dependent increase in efficiency.

This result has generated spirited debate. For example, the preceding commentary [4] contains familiar arguments; the gist being that enhancing quantum efficiency by breaking detailed balance somehow violates thermodynamics and/or commonsense. Space does not allow us to respond to [4] point by point; however, such a discussion will be presented elsewhere [5]. Here we present a detailed analysis of the simple laser based solar energy converter of Fig. 1, which will (hopefully) convince the reader that it is possible to increase quantum efficiency via quantum coherence. To that end, we next analyze the simple laser based solar energy converter of Fig. 1. As discussed in [6], a thermally pumped laser is a quantum heat engine. An incoherent thermal pump serves as the energy source at T_h that populates the upper laser level. The lower laser level is coupled to the ground state by "cold" light at T_c which serves as an entropy sink. The quantum efficiency is then $\hbar\nu_\ell/\hbar\nu_s = (1 - T_c/T_h)$.

Let us consider the solar pumped laser of Fig.1 in which the lower level doublet has a small amount of coherence such that $|\rho_{bc}| << \sqrt{\rho_{bb}\rho_{cc}}$. As is shown in detail in [3], the laser field $\mathcal{E}$ is now governed by

$$\dot{\mathcal{E}} = \kappa(2\rho_{aa} - \rho_{bb} - \rho_{cc} - \rho_{bc} - \rho_{cb})\mathcal{E} \tag{1}$$

where κ is an overall constant. To a good approximation, the populations are determined by T_h and T_c.

At threshold, Eq (1) implies that $1 - \rho_{bb}/\rho_{aa} - (\rho_{bc} + \text{c.c.})/2\rho_{aa} = 0$ where we have used the fact that $\rho_{bb} \cong \rho_{cc}$. Noting that $\rho_{bb}/\rho_{aa} = \exp - [\epsilon_b/kT_c - \epsilon_a/kT_h]$, the threshold condition then yields $\hbar\nu_s/kT_h - \hbar(\nu_s - \nu_\ell)/kT_c = ln[1 - (\rho_{bc} + \text{c.c.})/2\rho_{aa}]$, where in the notation of Fig. 1, $\epsilon_a = \hbar\nu_s$ and $\epsilon_b \cong \epsilon_c = \hbar(\nu_s - \nu_\ell)$. Finally we take $\rho_{bc} = |\rho_{bc}|e^{i\pi}$ and for weak coherence obtain

$$\hbar\nu_\ell/\hbar\nu_s = \eta_c + \delta\eta \ , \tag{2}$$

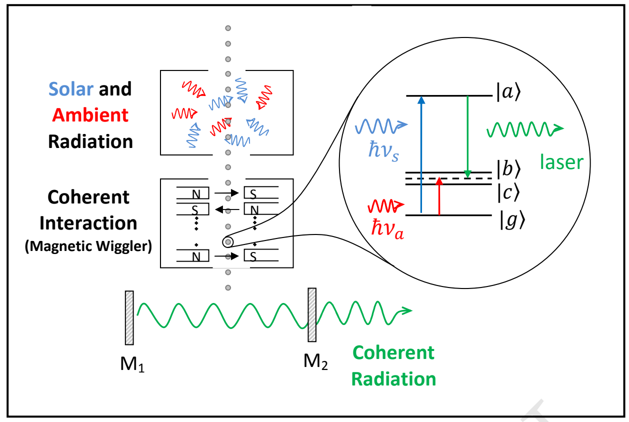

FIG. 1: Atoms pass through a beam of "hot" solar photons while interacting with ambient "cold" light at the same time. The population of upper level $|a\rangle$ and the lower level doublet $|b\rangle, |c\rangle$ is governed by Boltzmann factors characterized by temperatures T_h and T_c respectively. Then the atoms pass through a region where coherence ρ_{bc} is generated via, e.g., a wiggler array, etc. Finally the atoms enter into the laser cavity where coherent laser light (tuned to the midpoint between $|b\rangle$ and $|c\rangle$) is generated.

where $\delta\eta = kT_c|\rho_{bc}|/\hbar\nu_s\rho_{aa}$. Thus the conversion of incoherent solar photons to coherent laser photons (useful work) is enhanced by the factor $\delta\eta$. The connection between Eq (2) and the similar result in [2] is that in the present laser solar converter, the quantum efficiency is $\hbar\nu_\ell/\hbar\nu_s$ while in the semiconductor solar converter this efficiency is $eV/\hbar\nu_s$.Further analysis based on the previous quantum coherent photocell of ref [2] will be published elsewhere [5].

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