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Citation: *AIP Advances* **7**, 045004 (2017); doi: 10.1063/1.4979608

View online: <https://doi.org/10.1063/1.4979608>

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Power conversion efficiency of non-equilibrium light absorption

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(Received 2 December 2016; accepted 20 March 2017; published online 4 April 2017)

We deduce a novel expression for the non-equilibrium photochemical potential and the power conversion efficiency of non-equilibrium light absorption by a thermostated material. Application of our results for the case of electron migration from valence to conduction bands in photovoltaic cells allows us to accurately interpolate experimental results for the maximal efficiencies of Ge-, Si-, GaAs-based cells and the like. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). [<http://dx.doi.org/10.1063/1.4979608>]

Radiation-matter interaction is an ubiquitous and very important phenomenon in several fields of science including physics, chemistry, biology, medicine and technology.^{1–7} The key role of this interaction is that it promotes the absorption of the energy contained in the radiation that, in turn, may be converted and stored into other types of energy such as chemical or electrical. Radiation allows for an accurate control of nanomachines like information processing nanodevices, mobile biosensors, and regulators of chemical reactions and molecular assembly.^{1,8} The processes involving heat exchange by means of (non-thermal) radiation between materials at different temperatures are out of equilibrium. Therefore, beyond the Planck's seminal work on the thermodynamics of thermal radiation,⁹ one has to consider the existence of a non-vanishing photochemical potential associated to the photon-matter interaction.^{10,11}

Here, we analyze the radiative energy-exchange of two materials, α and β , which are thermalized with their corresponding heat baths at temperatures, $T_\alpha \neq T_\beta$, see Fig. 1. The kinetics of this non-equilibrium situation coupling the two materials is studied on the basis of a quantum master equation for two-level atoms, from which we deduce an explicit expression for the non-equilibrium photochemical potential of photons and the entropy produced during photon absorption. In energy conversion devices, like PV-cells, light absorption promotes the transference of electrons from valence to conduction bands. We prove that the "ultimate efficiency" factor limiting this conversion process can be calculated from the entropy production. Thus, our results constitute a generalization of the classical work by Schockley and Queisser (SQ) in which a radiation-matter thermal equilibrium is tacitly assumed when the detailed balance relation is used, see Refs. 7, 12. Previous works considering the implications that a solar cell is operating at steady-state conditions rather than in equilibrium can be summarized in the following references 13–16. Here, we start by considering the balance of radiative energy exchange between two systems of two state atoms thermalized at different temperatures in terms of quantum master equations for the state populations. These equations contain the matrix elements of the Hamiltonian of the matter-radiation system and therefore come, in principle, from the Von Neumann equation satisfied by the quantum density operator of the system. This approach is then used in a mesoscopic version of the non-equilibrium thermodynamics appropriate to this case due to the probabilistic nature of the description and the presence of energy and population fluctuations.¹⁹

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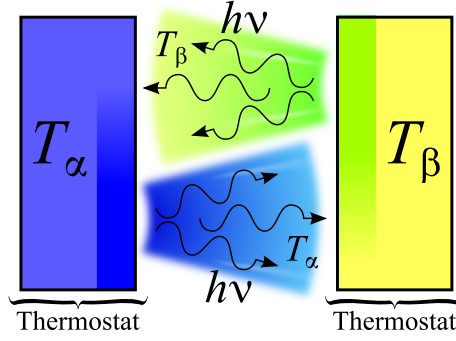


FIG. 1. Schematic representation of the system considered. Two sets of atoms, α and β in thermal contact with their respective thermostats at different temperatures T_α and T_β . Both materials interact with each other via radiation, $h\nu$.

In this form, our analysis starts by considering that each material is well represented as a system of two-level atoms in which transitions occur between their corresponding ground m to excited n states with energies E_m and E_n . These transitions match stimulated and spontaneous emission and absorption of photons of frequency $h\nu = E_n - E_m$. Mathematically, the model can be assembled with the help of Einstein's-like model^{17,18} for which the evolution equations for the normalized number of atoms in the ground state, m_i ($i = \alpha, \beta$) of materials α and β are

$$dm_i/dt = - \left[B_{mn}^i u_\nu^j + H_{mn}^i \right] m_i + \left[B_{nm}^i u_\nu^j + A_{nm}^i + H_{nm}^i e^{h\nu/k_B T_i} \right] n_i \quad (1)$$

Here, $j \neq i$ and n_i is the number of atoms in the excited state. B_{mn}^i and B_{nm}^i stand for the transitions probabilities per unit time associated to the stimulated radiation process and satisfying the detailed balance condition $B_{mn}^i = B_{nm}^i$. The spontaneous transitions rates from n to m are represented by A_{nm}^i . The thermal interaction of the atoms with the thermostats is quantified by means of the coefficients H_{nm}^i and H_{mn}^i , that satisfy the detailed balance relation $H_{nm}^i/H_{mn}^i = e^{h\nu/k_B T_i}$.¹⁸ Considering this last interaction is one of the main differences between our approach and that of Ref. 7.

The coupling between these two materials at different temperatures acts as a drift that takes the system away from equilibrium. Hence, it is convenient to rewrite Eq. (1) in terms of the difference between electromagnetic energy densities: $(u_\nu^j - u_\nu^i)$. Thus, by adding and subtracting terms in Eqs. (1), we may define the radiative currents

$$j_i \equiv -B_{mn}^i (u_\nu^j - u_\nu^i) (n_i - m_i), \quad (2)$$

and write the more compact equations

$$dm_i/dt = -j_i - k_i^+ m_i + k_i^- n_i. \quad (3)$$

Eq. (3) describes a first order chemical reaction without detailed balance, where k_i^+ and k_i^- are the corresponding forward and reverse rates constants or probabilities per unit time of passing from ground to excited states and viceversa. These constants are given by

$$k_i^+ \equiv B_{mn}^i u_\nu^j + H_{mn}^i, \\ k_i^- \equiv B_{nm}^i u_\nu^i + A_{nm}^i + H_{nm}^i e^{h\nu/k_B T_i}. \quad (4)$$

In Eq. (2), j_α is proportional to the net radiation received by material α from material β that promotes the transference of atoms from the ground to their excited state. In correspondence, j_β is proportional to the net radiation received by material β from material α . In equilibrium, $T_\alpha = T_\beta$ and thus j_i vanishes. In these conditions detailed balance is satisfied⁷

$$n_i^{eq}/m_i^{eq} = k_i^+/k_i^- = e^{-h\nu/k_B T_i}, \quad (5)$$

and, from Eq. (3) it may be derived the Planck's radiation formula¹⁸

$$u_\nu^j = \frac{8\pi h\nu^3}{c^3} \frac{\rho_j(\nu)}{e^{h\nu/k_B T_j} - 1}. \quad (6)$$

Here, we included the spectrum of the incoming radiation to material i will depend on the temperature and the emission properties of material j and viceversa. Thus, the overlapping of the absorption/emission properties of both materials will determine the specific frequencies that are permitted by the radiative interaction. This is accounted in Eq. (6) by the emission coefficient $\rho_j(\nu)$.

When $T_\alpha \neq T_\beta$ the entire system is out of equilibrium since there is a net flux of the number of atoms that perform transitions from their ground states m to their excited states n . Thus, Eq. (5) should be modified by considering a macro-canonical correction term accounting for the photochemical affinity per photon of frequency ν of material i , that is, the photochemical potential μ_i :

$$n_i(t)/m_i(t) \equiv e^{-h\nu/k_B T_j} e^{\mu_i/k_B T_j}, \quad (7)$$

where we have to emphasize that the radiation at temperature T_j acts as a heat bath for material i and viceversa.

The photochemical potential can be deduced by assuming stationary conditions in Eq. (3), that is, $dm_i/dt = 0$:

$$j_i = -k_i^+ m_i + k_i^- n_i. \quad (8)$$

Using now Eq. (2) we can obtain an expression for n_i/m_i :

$$\frac{n_i}{m_i} = \frac{1 + B_{nm}^i (u_\nu^j - u_\nu^i) / k_i^+}{1 + B_{nm}^i (u_\nu^j + u_\nu^i) / k_i^-} e^{-\frac{h\nu}{k_B T_i}}. \quad (9)$$

which can be substituted into Eq. (7) yielding the photochemical potential

$$\frac{\mu_i}{k_B T_j} = \frac{h\nu}{k_B} \left(\frac{1}{T_j} - \frac{1}{T_i} \right) + \ln \left| \frac{1 + \frac{B_{nm}^i}{k_i^+} \Delta u_\nu^{ji}}{1 + \frac{B_{nm}^i}{k_i^-} \Delta u_\nu^{ji}} \right|, \quad (10)$$

where $\Delta u_\nu^{ji} = u_\nu^j - u_\nu^i$ was introduced for notation convenience. In a first order approximation the expression of the photochemical potential is

$$\frac{\mu_i}{k_B T_j} \approx \frac{h\nu}{k_B} \left(\frac{1}{T_j} - \frac{1}{T_i} \right) + \chi_i (u_\nu^j - u_\nu^i), \quad (11)$$

where the frequency-dependent photochemical response function χ_i is defined by

$$\chi_i = \frac{B_{nm}^i (e^{h\nu/k_B T_i} - 1)}{A_{nm}^i + H_{nm}^i e^{h\nu/k_B T_i} + B_{nm}^i u_\nu^i}. \quad (12)$$

Including higher order terms of the series expansion in (11) yields a response function χ_i that depends in a non-linear way on the drift $u_\nu^j - u_\nu^i$. Other approaches introducing the concept of photochemical potential have been reported in the literature, see Refs. 10, 11. However, unlike the one developed here, out-of-equilibrium effects and the related dissipation are not considered.

For equilibrium processes, the *thermodynamic efficiency* η of a system performing irreversible processes is defined in a very general way by the well known expression: $\eta = W_{net}/Q_{in}$, in which W_{net} is the maximal available work and Q_{in} is the heat absorbed by the system in order to perform work. For irreversible processes it is more convenient to introduce the *power conversion efficiency*

$$\epsilon \equiv T\Sigma/\dot{Q}^{in}, \quad (13)$$

that introduces the rate of energy dissipated ($T\Sigma$) by the system in terms of the entropy produced per unit time Σ and \dot{Q}^{in} , the incoming energy per unit time. Defining the efficiency by using $T\Sigma$ is a direct way to measure the number of activated transitions that dissipate photon energy. This dissipated photon energy is used to produce hole-electron pairs in the valence and conduction bands. Hence, $T\Sigma$ is the maximal output power, i.e., the work which is transformed in an electronic current.

In this form, if $T_\beta > T_\alpha$, then the efficiency in the radiation absorption corresponds to material α and the *power conversion efficiency* ϵ can now be defined by

$$\epsilon_\alpha \equiv T_\beta \Sigma_\alpha / \dot{Q}_\alpha^{in}. \quad (14)$$

This quantity has its correspondence with the “ultimate efficiency” computed for PV-cells in Ref. 7. We emphasize that ϵ_α contains the temperature of the β -material also through the term \dot{Q}_α^{in} . There are other issues limiting the real efficiency of PV cells in practice: the carrier lifetime and diffusion length, defects, excitons, band alignment, interface properties and so on. All these factors bring about a lower efficiency and could properly be accounted for by following, for instance, the heuristic factors considered in Ref. 7. In our approach, some of these factors are implicitly incorporated through the coefficient H_{nm} that quantifies the interaction with the thermostat. For the explicit evaluation of ϵ_α we use Eq. (2) that introduces the incoming radiative energy flow

$$\dot{Q}_\alpha^{in} = \sigma T_\beta^4 j_\alpha \quad (15)$$

where σ is the Stefan-Boltzmann constant. The rate of entropy production of material α can be computed from the expression of Gibbs entropy¹⁹

$$S_\alpha = -k_B \sum_{a=m_\alpha, n_\alpha} a \ln \left(\frac{a}{a^{eq}} \right) + S_{eq}. \quad (16)$$

Hence, the rate of change of the entropy is given by the time derivative of (16): $dS_\alpha/dt = \psi_\alpha + \Sigma_\alpha$, where

$$\psi_\alpha = k_B j_\alpha \ln \left(\frac{k_\alpha^+ m_\alpha}{k_\alpha^- n_\alpha} \right), \quad (17)$$

is the entropy exchange due to the radiative interaction between materials and the second term

$$\Sigma_\alpha = -k_B (k_\alpha^- n_\alpha - k_\alpha^+ m_\alpha) \ln \left(\frac{k_\alpha^+ m_\alpha}{k_\alpha^- n_\alpha} \right) \geq 0.$$

constitutes the irreversible entropy production per unit time in material α , Σ_α . This second term is always positive in agreement with the second law of thermodynamics.¹⁰ Since ψ_α arises from the radiative coupling between both materials, it vanishes when $T_\alpha = T_\beta$, that is, in equilibrium.

The logarithmic term in Eq. (17) can be rewritten as a function of $\chi_\alpha (u_v^\beta - u_v^\alpha)$ by using Eqs. (5), (7) and (11). In the stationary state, $dS_\alpha/dt = 0$, and thus $\Sigma_\alpha = -\psi_\alpha$ that implies $j_\alpha = (k_\alpha^- n_\alpha - k_\alpha^+ m_\alpha)$, and therefore

$$\Sigma_\alpha = k_B \chi_\alpha j_\alpha (u_v^\beta - u_v^\alpha). \quad (18)$$

Using Eqs. (14), (15) and (18) one finally obtains

$$\epsilon = k_B \chi_\alpha \frac{u_v^\beta - u_v^\alpha}{\sigma T_\beta^3}. \quad (19)$$

Our results can be applied to calculate the efficiency of light absorption and electron migration from valence to conduction bands in PV-cells, see Fig. 2. In this case, the lower state of the semiconductor material α corresponds to the valence band with population m_α and the upper state to the conduction band with population n_α .

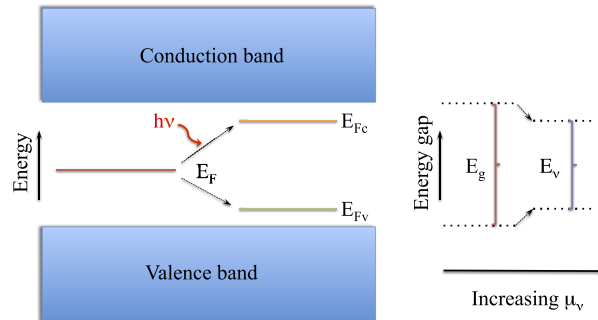


FIG. 2. Diagram of the bands structure of a typical semiconductor driven out-of-equilibrium by the action of an incident radiation field. Due to the radiation, the original Fermi level (E_F) splits into two quasi-Fermi levels E_{F_c} and E_{F_v} , in such a way that the equilibrium energy gap, E_g , is reduced to E_v .

In equilibrium, the energy E_{eq} of the semiconductor material α should be minimum. The incoming radiation takes the semiconductor away from equilibrium and therefore, the corresponding energy should be larger than in equilibrium: $E_{neq} > E_{eq}$. This is also true for the free energy per particle, that is, the chemical potential: $\mu_{neq} > \mu_{eq}$. The equality can be recovered if we add the chemical potential corresponding to the photons absorbed, that is, the photochemical potential (11). The resulting expression is

$$\mu_{neq} = \mu_{eq} + \mu_\nu. \quad (20)$$

The free energy can be recovered from Eq. (20) by noticing that the reaction coordinate ξ obeys the relations $d\xi = -dm = dn$, in agreement with the stoichiometric coefficients of Eq. (3). Integration of Eq. (20) with respect to ξ having in mind that μ_ν does not depends on ξ , yields

$$E_{neq}(\xi) = E_{eq}(\xi) + \mu_\nu \xi, \quad (21)$$

From this analysis, it follows that the photochemical potential tilts the equilibrium free energy, promoting in this way the transference of electrons from the valence to the conduction bands. At equilibrium, that is, for $\mu_\nu = 0$, the energy gap E_g between conduction and valence bands is given by

$$E_{eq}(n) - E_{eq}(m) = E_g. \quad (22)$$

When the system is taken away from equilibrium by radiation, that is, for $\mu_\nu \neq 0$, the Fermi level splits into two quasi-Fermi levels (see Fig. 2) separated by the open circuit voltage E_ν given by

$$E_{neq}(n) - E_{neq}(m) = E_\nu, \quad (23)$$

where the subindex ν stands for the fact that the splitting of the Fermi level is caused by the incoming radiation.

A comparison between experimental data²⁰ and the efficiency given by Eq. (19) can be performed in the case when the photon energy $h\nu$ coincides with E_g . Before discussing this comparison, in Fig. 3 we show the behavior of the power conversion efficiency as a function of the band gap energy for two radiation temperatures and χ_α given by Eq. (12). The figure emphasizes the important role of the coefficient H_{nm} in determining the maximal efficiency. This coefficient measures the thermal interaction of the atoms with the thermostat and other physical mechanisms that entail photon dissipation not associated to electron migration. The result shows that increasing the intensity of this interaction, that may be due to physical or construction imperfections among other factors

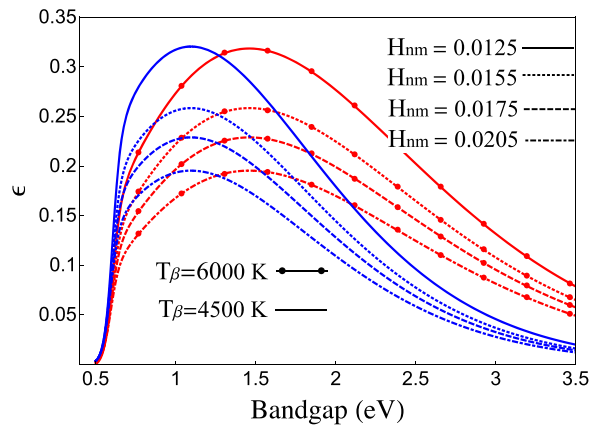


FIG. 3. Power conversion efficiency ϵ given by Eq. (19) as a function of the band gap E_g for two temperatures of the incoming radiation, $T_\beta = 6000$ K (red lines with circles) and $T_\beta = 4500$ K (blue lines). The different type of lines correspond to different intensities of interaction between the system and the heat bath, characterized by the parameter H_{nm} entering in the definition of χ_α in Eq. (12). The power conversion efficiency decreases when the parameter H_{nm} increases, indicating a higher energy dissipation to the heat bath at temperature $T_\alpha = 300$ K. The rapid decay of the efficiency at low eV's is due to the value of the spontaneous emission coefficient used, see the table of Fig. 4. For lower values of A_{nm} the curve becomes smoother at low eV's. The value of B_{nm} is also given in the table of Fig. 4.

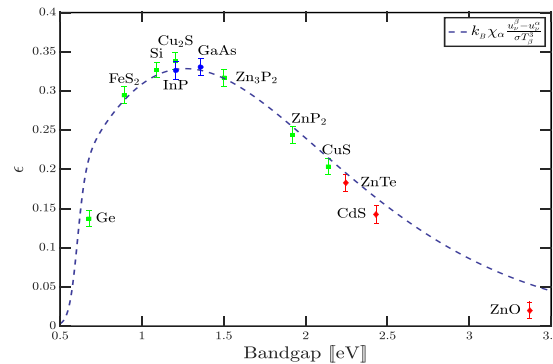
in practice, then the power conversion efficiency decreases. This is indicated by the solid, dotted, dashed and dotted-dashed lines of Fig. 3. Evidently, smaller values of H_{nm} entail greater efficiencies. The overall shape of the function depends on the value of the spontaneous emission coefficient A_{nm} . For values smaller than that used in Figs. 3 and 4 the function becomes smoother at low gap energies. However, the value used in these figures is consistent experimental data of PV-cells materials, see the [supplementary material](#).

The comparison of maximal power conversion efficiencies of different cells and the theoretical expression, Eq. (19), is shown in Fig. 4. A remarkable good interpolation of these maximal experimental efficiencies of several PV-cells taken from Ref. 20 was obtained, specially at low band gap energies. In doing this comparison we again used the microscopic expression of χ_α , Eq. (12). The table of values of the parameters A_{nm} , B_{nm} and H_{nm} is given in the same figure.²¹ See the [supplementary material](#) for a thorough explanation on the hypotheses regarding the deduction of the “ultimate efficiency” factor obtained by SQ.⁷ and its difference with the present approach. In the [supplementary material](#) we also explain how the physical data for the interpolation of experiments in Fig. 4 have been chosen.

As a summary, we have performed a thermostatical analysis of radiation-matter interaction which constitutes a case of an irreversible non-conductive heat exchange between materials at different temperatures. In particular, we have derived an explicit expression for the photochemical potential of the out-of-equilibrium radiation-matter interaction and the associated energy dissipation (entropy production). This has enabled us to give a general expression for the power conversion efficiency of the light absorption process, Eq. (19), and use it for accurately interpolate experimental results for the maximal efficiencies of Ge-, Si-, GaAs-based cells, see Fig. 4.

Our model is based on a master equation that, in principle, contains the matrix elements of the corresponding Hamiltonian of the matter-radiation system and therefore comes from the Von Neumann equation satisfied by the quantum density operator of the system, thus, it can be mapped onto first-principles. However, the final results are formally independent of the particular characteristic of the model and thus, are completely general. Our contribution, neither empirical nor semi-empirical, has no precedents in previous literature, since previous analyses of maximum efficiency did not take internal dissipation into account, see, for instance, Refs. 7, 12.

Our work offers a novel result with important technological implications in the field of energy harnessing, and may also be valuable in the realm of photochemical reactions in complex materials.



A_{nm} [s ⁻¹]	B_{nm} [m ³ J ⁻¹ s ²]	H_{nm} [s ⁻¹]	T_α [K]
1.5×10^8	5.5×10^{-3}	1.22×10^{-2}	300

FIG. 4. Data interpolation (curve), using Eq. (19), of the maximal experimental efficiencies (symbols) of several PV-cells taken from Ref. 20. In this case, we used the microscopic expression of χ_α , Eq. (12). The values of the parameters A_{nm} , B_{nm} and H_{nm} are given in the Table and correspond well to the values reported in Ref. 21 for Si. The maximum of the curve is matched by assuming a temperature $T_\beta = 5200K$.

SUPPLEMENTARY MATERIAL

See [supplementary material](#) for a thorough explanation of the concept of ultimate and power conversion efficiencies, and the election of the parameters for data interpolation in Fig. 4.

ACKNOWLEDGMENTS

This work was supported by UNAM DGAPA Grant IN116617, and by the MICINN of Spain under grant No. FIS2015-67837-P. APM acknowledges financial support by the Academic mobility program of the UNAM.

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