The Molecular Photo-Cell: Quantum Transport and Energy Conversion at Strong Non-Equilibrium

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The molecular photo-cell is a single molecular donor-acceptor complex attached to electrodes and subject to external illumination. Besides the obvious relevance to molecular photo-voltaics, the molecular photo-cell is of interest being a paradigmatic example for a system that inherently operates in out-of-equilibrium conditions and typically far from the linear response regime. Moreover, this system includes electrons, phonons and photons, and environments which induce coherent and incoherent processes, making it a challenging system to address theoretically. Here, using an open quantum systems approach, we analyze the non-equilibrium transport properties and energy conversion performance of a molecular photo-cell, including both coherent and incoherent processes and treating electrons, photons, and phonons on an equal footing. We find that both the non-equilibrium conditions and decoherence play a crucial role in determining the performance of the photovoltaic conversion and the optimal energy configuration of the molecular system.

Understanding the properties of non-equilibrium systems has been a central effort of the scientific community for many years. Of specific interest are non-equilibrium processes that take place at the nanometer scale and at which energy is converted from one form to another, for instance photovoltaic (PV) energy conversion, photochemistry and photosynthesis. In these cases, the interaction between electrons and photons under non-equilibrium conditions plays an essential role. Theoretical modeling of such processes is a challenging task, since the interacting nature of the system and its many-body characteristics, the multitude of constituents, the presence of external environments, and the non-equilibrium conditions must all be taken into consideration.

Even harder to address theoretically are situations in which the system has two independent fluxes, originating from separate non-equilibrium drivings, and are both far away from the linear response regime, a situation which is designated as strong non-equilibrium. A paradigmatic example for such a system is photo-voltaic cells, where the two fluxes are the heat flux, originating from the huge temperature difference between the sun and the earth, and the particle current originating from the bias voltage between the electrodes. In recent years a new and exciting class of photo-voltaic cells has emerged, namely molecular photo-cells, where the energy conversion process takes place at the single molecule level1–3.

Here we propose a formalism to study non-equilibrium transport in molecular junctions, and use it to investigate a model for the molecular photo-cell, a single molecular donor-acceptor complex attached to electrodes and subject to external illumination. This model was recently suggested4–6 (and a simpler version in Ref. 7) to be the minimal model to describe PV energy conversion in ideal, single-molecule heterojunction organic PV cells. In Refs. 4, 5 PV conversion efficiency was analyzed using the (essentially classical) rate equations for the electronic degrees of freedom. The dynamics and non-equilibrium properties of the phonons and photons were ignored, assuming to have equilibrium distributions. Here we show that the non-equilibrium properties of the phonons and photons have a strong impact on the PV conversion properties in realistic parameter range and cannot be neglected.

The formalism we present here allows us to treat electrons, photons and phonons fully quantum mechanically and on an equal footing (without resorting to a mean-field approximation) and to take into account the action of the environments producing a strong non-equilibrium situation. We use the many-body Lindblad quantum master equation8–10 to describe the environments, which consist of metallic electrodes in touch with a molecular
complex, a phonon bath (at ambient temperature) and a photon bath (at the solar temperature). Non-equilibrium is induced by two sources, namely the temperature difference between the incoming photons (originating from the sun) and the phonons with ambient temperature, and the bias voltage between the electrodes.

Using this formalism, we calculate the non-equilibrium densities of electrons, photons and phonons, the electric current and power output, and the thermodynamic efficiency at maximal power. We find that under certain conditions, the distribution functions for the phonons and photons can be very different from the equilibrium distributions, and therefore approximating the system as close to equilibrium is not a valid approximation. We then study the signature of non-equilibrium on the energy conversion efficiency of the system.

In addition to be able to include strong non-equilibrium effects and to account for all constituents on equal footing, our formalism allows one to introduce effects of dephasing and decoherence in a simple and physically transparent way. We study how the loss of quantum coherence affects the efficiency, and show that classical electron transfer (as opposed to coherent electron propagation) enhances the efficiency.

**Results: efficiency for the coherent system far from equilibrium**

The system under consideration is composed of a molecular junction, in which a donor-acceptor (D-A) complex is placed between two metallic electrodes (Fig. 1). This is an idealization of an environment, in which a donor-acceptor (D-A) complex is placed between two electrodes (Fig. 1). This is an idealization of an environment, in which a donor-acceptor (D-A) complex is placed between two metallic electrodes (Fig. 1). This is an idealization of an environment, in which a donor-acceptor (D-A) complex is placed between two metallic electrodes (Fig. 1). This is an idealization of an environment, in which a donor-acceptor (D-A) complex is placed between two metallic electrodes (Fig. 1). This is an idealization of an environment, in which a donor-acceptor (D-A) complex is placed between two metallic electrodes (Fig. 1). This is an idealization of an environment, in which a donor-acceptor (D-A) complex is placed between two metallic electrodes (Fig. 1).

The system consists of a molecule donor and an acceptor molecule, characterized by their HOMO and LUMO levels and coupled to each other via electron hopping. The D-molecule is coupled only to the left electrode, and the A-molecule only to the right electrode. Electrons in the donor interact with both photons (wiggly line) and phonons (broken line). The reader is referred to the Methods section and supplementary materials for a detailed description of the model and calculation. Recent advances in the experimental ability to measure photo-conductivity and PV conversion in single-molecule junctions make our theoretical model experimentally relevant.

We begin by examining the electron, photon and phonon densities at zero bias voltage. Note that the system is still out of equilibrium due to the temperature difference between the photon and phonon baths. In Fig. 2(a) steady state averages of the photon density \( n_{pht} = \langle \hat{n}_{pht} \rangle \), the phonon density \( n_{phn} = \langle \hat{n}_{phn} \rangle \), and the electronic occupation of the D-LUMO, \( n_{DLUMO} = \langle \hat{n}_{DLUMO} \rangle \), are plotted as a function of \( \epsilon_{-phn} \). Inset: typical current \( I \) and output power \( P_{out} = I \times V \) vs. bias voltage \( V \).
temperature is not applicable outside of equilibrium, it is still useful to think in terms of an effective temperature): the junction is locally (and efficiently) heated by the photons. This heat is transferred to the phonons, resulting in an elevated effective phonon temperature and, consequently, enhanced phonon occupation. (iii) When e–photon coupling is large, \( \lambda_{\text{ph}} \gg \lambda_{\text{phn}} \), there is no longer efficient transfer of heat to the phonons. However, the D-LUMO occupation continues to rise, due to energy pumping from the photons to the D-LUMO.

An important measure of the operational efficiency of the molecular PV cell is the efficiency at maximal power, \( \eta_{\text{max}} \), defined in the ratio between the cell’s maximal output power \( P_{\text{out}} \) and the corresponding input power \( P_{\text{in}} \) supplied by the photons\(^{4,5}\). The output power is given by \( P_{\text{out}} = J \times V \), where \( J \) is the current through the system, and the input power is calculated in a similar way\(^{6} \)

\[
P_{\text{in}} = 2 \alpha \omega_{\lambda} \lambda_{\text{ph}} \left( a_{D1}^* c_{D1} + a_{D2}^* c_{D2} - a_{A1} c_{A1} - a_{A2} c_{A2} \right)
\]

which is related to the photon-induced part of the particle current.

The inset of Fig. 2(b) shows a typical \( J \) vs \( V \) characteristics (blue) and the bias voltage \( V \) dependence of the output power (purple). As seen in the figure, there exists a bias voltage \( V_{\text{max}} \) which gives the maximum efficiency. In Fig. 2(b), the efficiency at \( V_{\text{max}} \), i.e., the efficiency at maximal power, is plotted as a function of the \( e – \text{photon} \) coupling (we use the same parameters as in Fig. 2(a)). For very small \( e – \text{photon} \) coupling (\( \lambda_{\text{ph}} < 0.002 \text{ eV} \)), the efficiency is very small, and grows linearly with \( \lambda_{\text{ph}} \). In this regime, the time it takes for an electron to absorb a photon is larger than the time the photons spent in the cell (defined by \( \gamma_{\text{ph}} \rho_n(T) \)).

In Fig. 3 we plot the efficiency at maximum power \( \eta_{\text{max}} \) as a function of the exciton Coulomb energy \( U \).

Results: the role of decoherence

The next question we wish to address is the extent to which the quantum nature of the system affects the PV conversion efficiency, a question which is beyond the reach of the formalism presented in Refs. 4, 5. The formalism we present here allows us to access, in addition to fully quantum-coherent processes described above, also incoherent processes. The most important incoherent processes are electron transfer from the D-LUMO to the A-LUMO, described classically in Ref. 4. These are addressed here by adding an additional pair of \( \hat{V} \)–operators that accounts for incoherent transitions, namely

\[
\hat{V}_{D-A} = \Gamma_{D-A} e^{D2} a_{D2}^* e^{A2} a_{A2} - \frac{1}{2} \Theta(0.5) \Theta(0.5) \Theta(0.5)
\]

We now parameterize \( \Gamma_{D-A} \) and \( \gamma_{D-A} \) with \( \xi \), \( \Gamma_{D-A} = \Gamma_{D-A}^{0.5} \Theta(0.5) \Theta(0.5) \Theta(0.5) \) (the Heaviside step-function) is the Heaviside unit step-function). This parameterization is shown on the right inset of Fig. 4, and it is constructed such that for \( \xi = 0 \), the system is fully coherent, for \( \xi = 0.5 \) the system is mixed (both quantum and classical processes), and for \( \xi = 1 \) the system is fully incoherent, so the region \( 0 < \xi < 1 \) interpolates between all three cases.

In Fig. 4 we plot the efficiency at maximum power \( \eta_{\text{max}} \) as a function of the position of the A-LUMO, \( \epsilon_A \), and the parameter \( \xi \). We set \( \gamma_{D-A} = 10^{+3} \text{ s}^{-1} \) as in Ref. 4. We find that the quantum coherence or classical decoherence (parameterized by \( \xi \)) has a profound effect on the efficiency of the molecular PV-cell in two important aspects.

First, the optimal position of the A-LUMO energy differs according to the nature of the transition under consideration: quantum (coherent), both quantum and classical, or classical D–A transitions (solid lines in Fig. 4). For the last case (\( \gamma_{D-A} = 0 \)), we find that \( \epsilon_A \) is optimal at \( \epsilon_A = 1.25 \text{ eV} \), verifying the result of Ref. 4. When quantum correlations are added (\( \Gamma_{D-A} = 0.05 \text{ eV} \), \( \gamma_{D-A} = 10^{+3} \text{ s}^{-1} \)), two peaks emerge at \( \epsilon_A \sim 0.9, 1.3 \text{ eV} \), and a lower peak emerges at \( \epsilon_A \sim 1.6 \text{ eV} \). For a system with only quantum transitions (\( \Gamma_{D-A} = 0.05 \text{ eV} \), \( \gamma_{D-A} = 0 \)), enlarged in the back inset in Fig. 4), the lower peak vanishes, and the optimal LUMO positions are at 1.2 eV and 1.6 eV. Thus, in the design of optimal molecular PV cells, it is important to take into account the quantum nature.
Second, as can be clearly seen in Fig. 4, the addition of classical D-A transitions increases the efficiency substantially by more than an order of magnitude. This finding is surprising, since one would expect that incoherent (and dissipative) transitions would lead to a decrease in the efficiency. To understand the origin of this effect, we performed time-dependent calculations (not shown) for a system composed of D-LUMO, acceptor, and the coupling with right electrode, and found that for the coherent case, an electron that is excited to the D-LUMO coherently oscillates between the D- and A-LUMO, while for the incoherent case the electron decays from the D- to the A-LUMO exponentially (and its return rate is exponentially small).

This implies that in the coherent case the electron spends much more time in the D-LUMO than in the incoherent case, before transferring to the right electrode. Since the electron can decay back to the D-HOMO (emitting a phonon) only directly from the D-LUMO, the longer it spends on the D-LUMO, the higher the probability for non-HOMO (emitting a phonon) only directly from the D-LUMO, the higher the probability for non-HOMO electron transfer processes. As can be seen, the decay rate is much smaller for the fully coherent case, indicating a longer relaxation time. This is in line with the observation above that slower relaxation dynamics lead to lower efficiency. In addition, we point out that the relaxation times of the D-A LUMOs and the right electrode (which are numerically much easier to calculate than the efficiency of the full system including D-HOMO, photons, phonons, and related Lindblad dissipators) serve as an indicator for the efficiency, even though they do not capture the fine details required for an optimal design of the system (see Fig. 4(a) and (b)).

To further examine possible effects of coherence on the efficiency of the molecular PV cell, we study a system where the donor has two degenerate D-LUMO levels which have been introduced experimentally[22]. Here, we study a simplified system (schematically depicted on the right side of Fig. 6), in which the photons and the phonons excite electrons with equal amplitudes from the D-HOMO to the two D-LUMO levels. Each of the levels is coupled to the A-LUMO with the same hopping amplitude $t_{D-A}$, and they are coupled to each other with a complex hopping amplitude $h$. In Fig. 6, the efficiency at maximum power $\eta_{mx}$ is plotted as a function of the inter-LUMO coupling $h$ (solid line) and the phase $\phi$ (dashed line) for $\gamma_{D-A} = 0$, i.e., no incoherent D-A transfer ($\gamma_{split} = 0.1$ eV). We find that while $h$ has little effect on the efficiency, the phase $\phi$ has a significant effect (increasing the efficiency by up to $\sim 15\%$).
indicating the effect of quantum coherence on the efficiency for fully coherent D-A transfer ($\gamma_{D-A} = 0$). The dotted line is the same for a mixed coherent-incoherent transfer ($\gamma_{D-A} \neq 0$).

Surprisingly, we also find that this quantum interference effect persists even when incoherent D-A transfer is included ($\gamma_{D-A} = 10^{-3}$ s⁻¹ as in Fig. 4), and an substantial increase of $\eta_{max} \sim 30\%$ is observed by varying $\phi$ (dotted line).

**Conclusions**

In Summary, we have proposed a novel formalism to study non-equilibrium quantum transport in molecular junctions, and applied it to investigate a minimal model of PV energy conversion in ideal, single-molecule PV cells. The results shown above indicate that quantum coherence effects are important in determining the non-equilibrium energy conversion performance of molecular PV cells. The formalism presented here sets the stage for a fully coherent quantum mechanical calculation of energy conversion in more realistic models for molecular PV cells and can be directly linked to quantum chemistry methods (such as density-functional theory). The progress in the experimental ability to measure photo-conductivity and PV conversion in single-molecule junctions allows one to envision real PV devices composed of a single molecular junction or a molecular monolayer, making our theoretical model experimentally relevant. Furthermore, our method can include both coherent and incoherent effects, making it a useful tool in the study of other energy conversion processes such as photosynthesis, where both classical and quantum processes take place, or other chemical and photo-chemical processes.

**Methods**

The full Hamiltonian of the molecular PV cell, including the molecular orbitals, the photons and the phonons, may be written as $H = H_M + H_{ph} + H_{phn} + H_{M-ph} + H_{M-phn}$, where $H_M$ is the Hamiltonian for the molecular complex, $H_{ph(phon)}$ is the photon (phonon) Hamiltonian, and $H_{M-ph(phon)}$ describes the electron-phonon (phonon) interaction (we set $\hbar = 1$ hereafter),

$$H_M = \sum_{q} \epsilon_q c_q^\dagger c_q - \mathbf{T}_{a} - \mathbf{A}(\mathbf{c}_q^\dagger c_q + H.c),$$

$$H_{ph} = \sum \epsilon_i a_i^\dagger a_i,$$

$$H_{phn} = \sum \epsilon_i b_i^\dagger b_i,$$

$$H_{M-ph} = \lambda_{ph} c_i^\dagger c_j b_j + H.c.,$$

$$H_{M-phn} = \lambda_{phn} b_i^\dagger c_j^\dagger c_i + H.c..$$

Here $\epsilon_q(c_q)$ creates (annihilates) an electron in the D-HOMO ($x = D, 1$), D-LUMO state ($x = D, 2$) or A-LUMO state ($x = A$), with the corresponding level energies $\epsilon_q$, $\tilde{\epsilon}(a)$ creates (annihilates) a photon with energy $\omega_a = \omega_D - \epsilon_D - c_{D,1}$, and $b_i^\dagger (b_i)$ creates (annihilates) a phonon with the same energy. In principle one should consider many photon (and phonon) modes, however the strongest effect on the dynamics comes from the resonant photons (with energy same as the HOMO-LUMO gap). The electron-photon Hamiltonian $H_{M-ph}$ describes (within the rotating wave approximation) the process (and its reverse process) in which an electron in the D-LUMO state relaxes to the D-HOMO state and emits a photon, with the electron-photon ($e-ph$) coupling $\lambda_{eph}$. The electron-phonon Hamiltonian $H_{M-phn}$ is similar to $H_{M-ph}$, but with phonons instead of photons. While spins might play a role in real systems (for example by introducing selection rules for allowed transitions), we chose to introduce a simplified (toy) model in which spins play no role in the transport processes.

To study the dynamics of the system, we use the Lindblad equation to model the system and the environments:

$$\dot{\rho} = -i[H, \rho] + \sum (\frac{1}{2} [\gamma_{ph} \rho + \gamma_{phn} \rho] + \gamma_{ph} \rho),$$

where $[\cdot, \cdot]$ is the commutator and $[\cdot, \cdot]$ is the anti-commutator.

The essence of the Lindblad approach is that instead of describing the environment by encoding it into a self-energy (as is done in the non-equilibrium Green's function approach), the environment is characterized by its action on the system. This action is mapped onto so-called Lindblad $\gamma$-operators, which describe incoherent transitions of the system elements due to the presence of an environment. The Lindblad equation was recently employed to address various aspects of electron transport, yet in these studies the interaction with an environment was limited to electrons only, and the non-equilibrium dynamics of other constituents (i.e. phonons or photons) was not considered.

We assume that the left electrode is coupled only to the D-HOMO and that the right electrode is coupled only to the A-LUMO, as in Fig. 1. The corresponding $\gamma$-operators are then:

$$\gamma_{LH} = \sqrt{\gamma_{ph} n_{L}(T)} a_i^\dagger a_i,$$

$$\gamma_{RH} = \sqrt{\gamma_{ph} n_{R}(T)} b_i^\dagger b_i.$$
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