Factors limiting the efficiency of molecular photovoltaic devices

Nelson, J.[a] , Kirkpatrick, J.[a] and Ravirajan, P.[ab]

[a] Ctr. for Electron. Mat. and Devices, Department of Physics, Imperial College London, United Kingdom
[b] Department of Physics, University of Jaffna, Jaffna, Sri Lanka

Abstract

We present a simple model of a molecular photovoltaic device consisting of a two-level system, connected to external contacts by chains of one or more charge transporting orbitals. Electrons may be promoted in the two-level system by photon absorption, and charge transported to the external circuit by electron transfer between neighboring orbitals. Photon absorption and emission are described by a generalized Planck equation and electron transfer is described by nonadiabatic Marcus theory. We find the steady-state current by solving the set of coupled rate equations for electron transfer in the system under illumination as a function of bias applied to the contacts. We calculate monochromatic current-voltage characteristics and power conversion efficiency as a function of the system size, orbital energy levels, and electron transfer rates, and compare with the monochromatic detailed balance limit. Using realistic values of the energy levels and charge-transfer rates, we are able to reproduce a number of commonly observed features in the current-voltage characteristics. These include a "kink" in the current-voltage curve close to open circuit when large interfacial energy steps are present or mobilities are low, and a reduction of the open-circuit voltage and crossing of the light and dark current curves when interfacial recombination is strong. We show that open-circuit voltage is dominated by the acceptor-donor energy gap when recombination is important, and by the optical gap when recombination is low. We confirm previous reports that photovoltaic energy conversion can be achieved by interfacial asymmetry alone and that a potential difference between the electrodes is unnecessary. Improved photovoltaic efficiency of molecular heterojunctions requires ohmic contacts, improved charge-carrier mobilities, and tuning of the electron-transfer rates at the heterojunction. Maximizing the rate of charge separation does not necessarily lead to maximum efficiency.

Indexed keywords

EMTREE medical terms: article; calculation; darkness; density; device; electrode; electron transport; electronics; energy; illumination; light absorption; molecular photovoltaic device; molecule; photon; steady state; temperature; velocity