

Optimization of Carrier Transport Processes and Figure of Merit for an Intermediate Band Solar Cell

Investigators

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Abstract

This exploratory research work is focused on the use of ultrafast terahertz spectroscopy as an all-optical probe of the first steps in carrier transport processes in energy related materials. First experiments have been carried out probing these processes in intermediate band materials. Intermediate-band materials have the potential to be highly efficient solar cells and can be fabricated by incorporating ultrahigh concentrations of deep-level dopants. We used optical-pump/terahertz-probe measurements to study carrier recombination dynamics of chalcogen-hyperdoped silicon with sub-picosecond resolution. The recombination dynamics is described by two exponential decay time scales: a fast decay time scale ranges between 1 and 200 ps followed by a slow decay on the order of 1 ns. In contrast to the prior theoretical predictions, we find that the carrier lifetime decreases with increasing dopant concentration up to and above the insulator-to-metal transition. Evaluating the material's figure of merit reveals an optimum doping concentration for maximizing performance. We have also very recently applied these techniques to co-doped intermediate band materials in which sulfur is used to hyperdope and form the intermediate band while boron is used to tune the Fermi energy within the intermediate band. These first measurements allow for direct probing of both the electron and hole dynamics and direct extraction of effective mobilities, a key factor in determining the efficiency of a IB solar cell. Finally, as a comparison set, we have also applied these same techniques to probe carrier mobilities in polymer-fullerene organic solar cell materials, probing the short time mobilities in regioregular and regiorandom P3HT:PCBM and the dependence of the mobility on the polymer side chain structure.

Introduction

The goal of this research is to investigate carrier recombination dynamics in intermediate band materials. Theoretical calculations show that an intermediate band solar cell can achieve an efficiency limit of 63%, equivalent to an ideal triple-junction cell, about 150% that of the Shockley-Queisser limit. So far, devices have showed broad band photocurrent response,[1] but failed to achieve these high efficiencies,[2] likely due to short carrier lifetimes. Long-wavelength electromagnetic radiation in the terahertz (THz) frequency is sensitive to free charge carriers, and sub-picosecond THz pulses can be generated by ultrafast lasers and nonlinear medium. Time-resolved terahertz spectroscopy enables us to map the carrier recombination dynamics on picosecond time scales. We measure electron recombination time-scales in chalcogen-hyperdoped silicon, where intermediate band is introduced by ultrahigh concentrations of chalcogen dopants (S, Se or Te), and show that there is an optimal dopant concentration at which the performance figure of merit is optimized.[3] Current effort is focused on evaluating both

the electron and hole transport dynamics in intermediate band materials since both high figure of merits for both charge carriers are required for a successful intermediate band solar cell.[4]

The second area currently under investigation is carrier transport dynamics in organic solar cells. Organic bulk-heterojunction solar cells are promising for achieving high efficiencies since photogenerated electrons transfer from the donor to the acceptor at a very fast rate. Record photoconversion efficiency is almost 10%, but at the same time the charge separation mechanism is not well understood. We use time-resolved terahertz spectroscopy to study frequency dependent carrier mobility. We compare THz mobility of regioregular poly(3-hexylthiophene) (P3HT) and regiorandom P3HT, where the side chains are oriented differently, and find the local polymer chain structure has a large impact on carrier mobility. Accurate measurement of short-time carrier mobilities will also shed light on how carrier mobilities reflect photoconversion efficiencies of these solar cells.

Background

Although terahertz spectroscopy has been widely applied to the study of carrier dynamics in materials, the measurements described above are the first THz-probe measurements carried out in intermediate band solar cell materials and enable for the first time direct measurements of the effective figure of merit. With respect to the studies of organic solar cells, there has been prior work in this area but a fundamental understanding of the short time mobility and its dependence on the local microscopic structure of the polymer has never been carried out.

Results

Intermediate band solar cell performance figure of merit

Figure 1a shows the electron figure of merit of sulfur hyperdoped silicon. Solar cell performance figure of merit indicates the material's potential for a successful intermediate solar cell. Figure of merit is given by

$$\text{Figure of merit} = \frac{E_g}{q} \alpha^2 \mu \tau, \quad (1)$$

Here E_g is the band gap energy, q is the elementary charge, α is the sub-band gap light absorption coefficient, μ is mobility of the carriers, and τ is the electron carrier life time measured in this work.[4] Figures 1b, 1c, and 1d show how each property changes with dopant concentration. Although figure of merit greater than 1 is desired for a useful intermediate band photovoltaic, our study shows that there is an optimal dopant concentration for the highest figure of merit, and this concentration is far below the insulator-to-metal transition (IMT) concentration, a concentration level previous theoretical works suggest intermediate band material should exceed.[5]

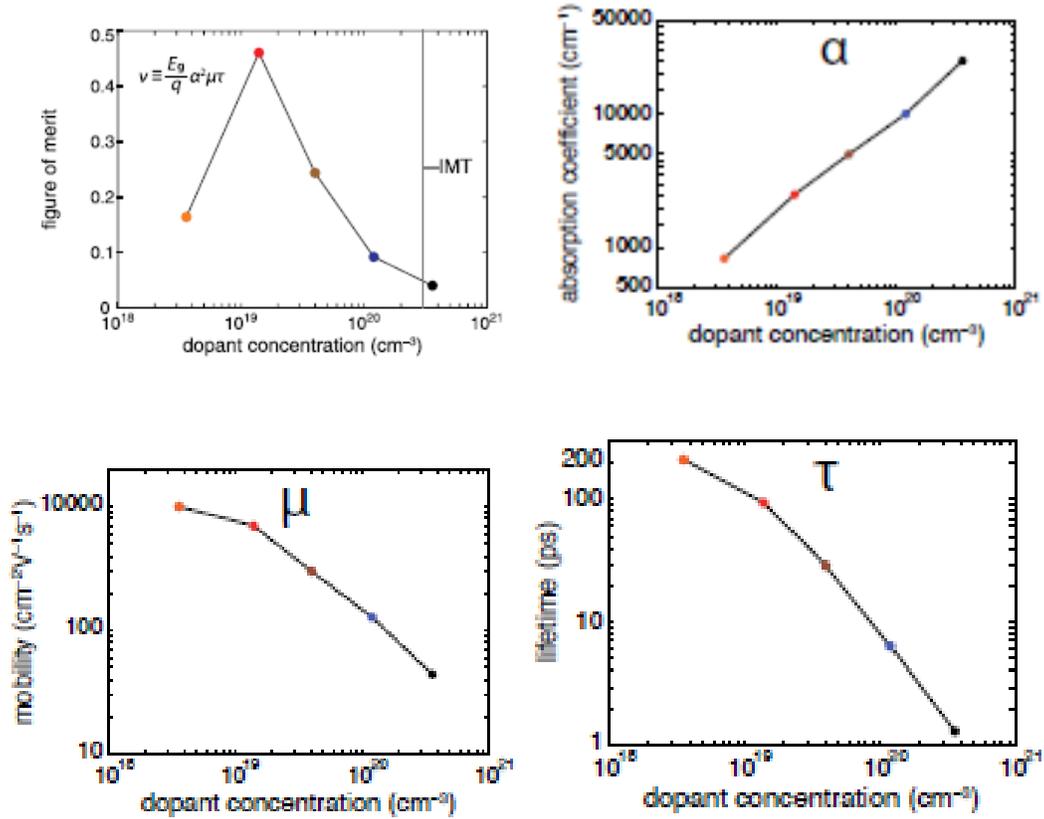


Figure 1: (a) Calculated figure of merit of S-hyperdoped Si samples. The sample with concentration above the IMT (vertical line) absorbs sub-band gap light most strongly but has the lowest figure of merit since both mobility and carrier lifetime are much lower. The sample with the highest figure of merit strikes a better balance between light absorption, mobility, and lifetime. (b) Literature values of sub-band-gap light absorption coefficient (at 0.5 eV) and (c) carrier mobility. (d) Carrier lifetime as a function of dopant concentration for both S-hyperdoped Si samples.

Terahertz Carrier Mobility in Polymer-Fullerene Organic Semiconductors

Figure 2 shows effective frequency dependent mobility of two organic polymer solar cell materials. We investigate how carrier mobility is impacted by the regioregularity of P3HT polymer side chains. In regiorandom P3HT:PCBM (Figure 2 inset), the random orientation of the polymer sidechain introduce more bends of the polymer backbone and hence the overall structure of the polymer:fullerene solar cell. Studies show that regioregular P3HT:PCBM solar cells exhibit higher power conversion efficiency (4%) than regiorandom P3HT:PCBM (2.5%), but the origin of the efficiency reduction is not well known.[6] Our work indicates that even at a local charge transport level, the side chain orientation has large impact on charge transport.

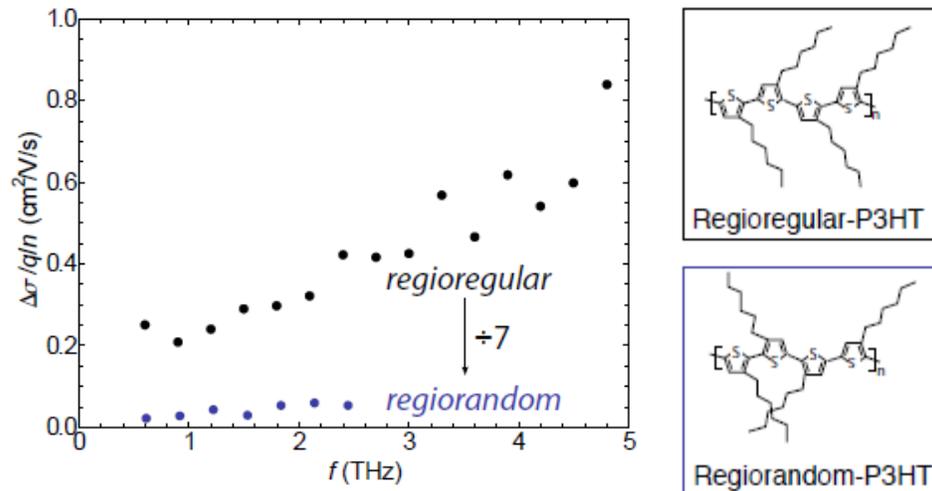


Figure 2: (a) Effective carrier mobility of P3HT:PCBM (1:1) as a function of THz frequency. (b) and (c) schematics of the P3HT polymer back bone and side chain structure.

Progress

Within this exploratory proposal in its first half year, it is probably too early to say what the direct impact on reduction in emissions of greenhouse gases will be. However if successful this work will enable a new feedback process by which intermediate band solar cell materials can be quickly characterized through direct measurement of their figure of merit.

Future plans

Completion of studies on co-doped IB materials, in particular investigating both electron and hole carrier dynamics by tuning the Fermi level within the intermediate band device. Completion of studies on organic polymer solar cells.

Publications and Patents

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