

Charge transport and recombination in MDMO-PPV/PCBM bulk heterojunction solar cells probed by the photoinduced charge extraction by linearly increasing voltage technique

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The high quantum yield of charge generation within the interpenetrating network of electron donating conjugated semiconducting polymers and electron accepting fullerenes is a prerequisite for efficient photovoltaic cells, yet the charge collection efficiency depends on the charge carrier mobility and the lifetime of the photogenerated charge carriers.

The mobility and recombination of the charge carriers is *simultaneously* studied within the MDMO-PPV/PCBM mixture using the photoinduced charge extraction by linearly increasing voltage technique (Photo-CELIV). The sample, which is typical of a bulk heterojunction solar cell, consists of a thin (200 -300 nm) layer of the photoactive blend sandwiched between a transparent ITO-coated glass and evaporated aluminum electrodes. Upon application of a reverse bias triangular-shaped voltage pulse with a voltage rise speed $A=dU/dt$, the typical electrical response is a rectangular-shaped current transient with a plateau value corresponding to the capacitive displacement current $j(0) = A \times \epsilon \epsilon_0 / d$, where ϵ , ϵ_0 is the dielectric constant of the material and vacuum, respectively, and d is the thickness of the dielectric (see Fig. 1 left). When a strongly absorbed laser flash hits the sample, charge carriers are photogenerated throughout the layer. The photogenerated charge carriers either undergo recombination or exit the device through the external circuit under the influence of the built-in electric field (short circuit condition). The external photocurrent upon photoexcitation can be minimized by the application of a DC offset bias ($U_{offset} = U_{bi}$) leading to flat bands and forcing the charge carriers to recombine. The remaining charges can be extracted after an adjustable delay time (t_{del}) determining their lifetime, meanwhile their mobility (μ) can be calculated according to Eq. 1:

$$\mu = \frac{2d^2}{3At_{max}^2 \left[1 + 0.36 \frac{\Delta j}{j(0)} \right]} \quad \text{if} \quad \Delta j \leq j(0) \quad (1)$$

where Δj is the extraction current maximum and t_{max} is the time to reach this maximum extraction current.

The Photo-CELIV transients recorded at room temperature at (a) various delay times between the light pulse and the extraction pulse, and (b) at various light intensities at a fixed delay time are shown in Fig. 1 (right). Interestingly, t_{max} shifts to longer times as the delay time between the light pulse and the extraction pulse is increased, which is related to a decreasing mobility according to Eq. 1. Moreover, the maximum of the extraction current (Δj) decreases, which is related to the recombination of the charge

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carriers. The t_{\max} remains almost constant when the light intensity is decreased at a fixed delay time (Fig. 1 right (b)), and the number of extracted charge carriers saturates with increasing light intensity above $\sim 1 \mu\text{j}/\text{pulse}/\text{cm}^2$.

The inset of Figs. 1 a and b show the dispersion of the Photo-CELIV transients characterized by $t_{1/2}$ to t_{\max} . The recorded Photo-CELIV transients are dispersive at short delay times, which gradually decrease to the ideal non-dispersive value of 1.2 at longer time delays. On the contrary, the $t_{1/2}$ to t_{\max} values do not change significantly by varying the light intensity (charge carrier concentration) indicating a concentration independent but time dependent dispersion at short delay times. Finally, the majority of the photogenerated charge carriers can be extracted by the extraction pulse, which indicates that trapping into deep electronic traps at this time scale is negligible.

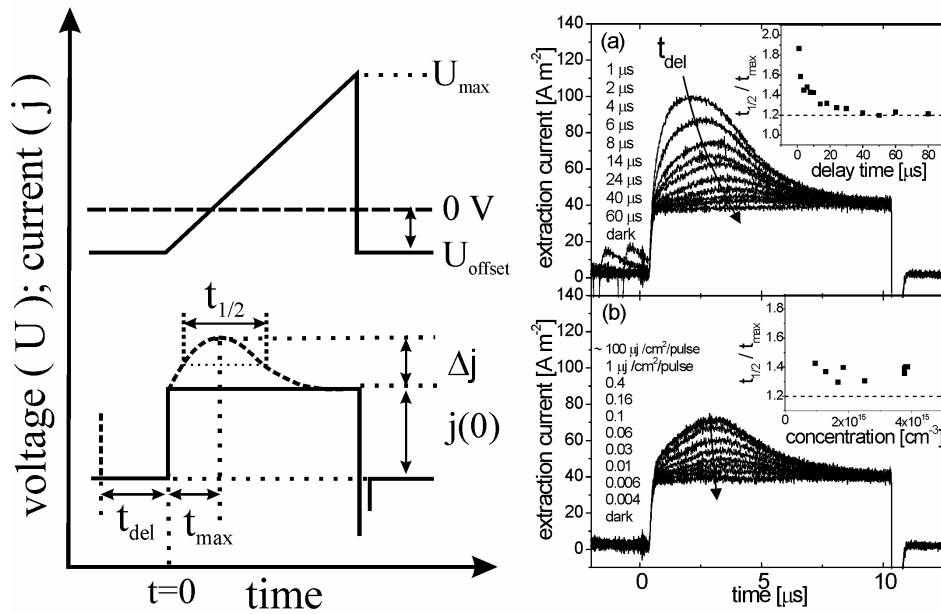


Fig. 1 (Left) Schematic response and the timing sequence of the Photo-CELIV technique. (Right) Photo-CELIV transients recorded for a ITO/MDMO-PPV:PCBM (1:4)/Al device at (a) various t_{del} (b) various light intensity. The inset shows the dispersion parameters defined as $t_{1/2}$ to t_{\max} .

The charge carrier mobility at room temperature is $\sim \mu=2 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which slightly decreases with increasing the active layer thickness. The recombination of the charge carriers is studied by plotting the concentration of extracted charge carriers versus delay time, and a hyperbolic decay typical of a bimolecular recombination law is found.

Finally, the Photo-CELIV transients have been recorded versus delay time, light intensity and applied electric field at several temperatures. These studies indicate an increasing dispersion (time-dependence) of the mobility and correspondingly, the recombination dynamics of the charge carriers. A dispersive bimolecular recombination model is presented, which can properly describe the observed concentration decay of the photoinduced charge carriers at all temperatures and applied delay times. The relevance of the measured mobility and lifetime values to operational conditions is evaluated by analyzing the current density versus voltage curves of bulk heterojunction solar cells with varying active layer thickness.