

Transient Hole Transport in Poly(*p*-phenylene vinylene) LEDs

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Abstract

The transient transport of holes in poly(*p*-phenylene vinylene) (PPV) is studied experimentally by monitoring the response times of PPV based light-emitting diodes (LEDs). For thin LEDs, a dispersive hole current is observed. This is well described by continuous-time random walk theory. It appears that the dispersion in hole transit times is mainly due to structural disorder in the polymer layer. For thicker LEDs, a transition towards nondispersive hole transport is observed. In this regime, the response times correspond well with the hole mobility observed in stationary experiments. The experimental results are compared with theoretical estimates for the transition from dispersive to nondispersive transport.

Keywords: Transport measurements, Poly(phenylene vinylene) and derivatives, Light sources.

1. Introduction

Conjugated polymers, particularly poly(*p*-phenylene vinylene) (PPV) and its derivatives, have been a subject of intense research over the last decade because of the potential application in polymer light-emitting diodes (LEDs) [1]. Current density-voltage (*J-V*) measurements on PPV-based LEDs and on hole-only devices (where the electron current has been blocked) have revealed that the current is dominated by holes. This hole current is consistently described as a function of electric field *E*, temperature *T*, and layer thickness *L* by a combination of space charge effects and a temperature and field-dependent hole mobility of the form [2]

$$\mu_p = \mu_0 \exp \left[-\frac{\Delta}{k_B T} + B \left(\frac{1}{k_B T} - \frac{1}{k_B T_0} \right) \sqrt{E} \right]. \quad (1)$$

Typical parameter values are $\Delta=0.5$ eV, $B=3 \times 10^{-5}$ eV (V/m)^{-1/2}, and $T_0=600$ K. This form of the direct current (DC) mobility has been found in numerous disordered organic semiconductors [3] and is thought to arise from hopping transport in a system with both energetic and structural disorder [4].

Recently, we have studied the transient hole transport characteristics [5] using the delayed electroluminescence (EL) technique [6,7]. This technique is based on the notion that the delay between a voltage pulse and the onset of EL in a polymer LED is directly related to the transit time of holes towards the cathode. The advantage of this technique is that it can be used for layers with a thickness of the order of 100 nm, which is about an order of magnitude thinner than in conventional time-of-flight (TOF) experiments.

We have found that the EL delay time is not determined by the hole mobility [Eq. (1)], which characterizes the *average* hole

transit time, but by the *dispersion* in hole transit times, in particular by the transit time of the fastest holes. At room temperature, the delay time τ_d follows the universal scaling law $\tau_d \sim (L/E)^{1/\alpha}$, in agreement with the continuous-time random walk theory by Scher and Montroll [8]. Here, the dispersion parameter α ($0 < \alpha < 1$) is a measure for the distribution of waiting times between successive hops. The “mobilities” that are obtained from these transit times follow from

$$\mu_p \equiv \frac{L}{\tau_d E} \sim \left(\frac{E}{L} \right)^{(1-\alpha)/\alpha}. \quad (2)$$

Studying τ_d as a function of temperature, we found that the average activation energy of a hop amounts to $\Delta_0=0.34$ eV, and that $\alpha=0.45$, independent of temperature. Since energetic disorder would give rise to a temperature dependent α , we concluded that structural disorder is the main contributor to the dispersion in hole transit times [5].

This result seems to be at variance with the recent TOF measurements on PPV by Lebedev *et al.* [9], who interpreted the power-law decay of the transient photocurrent in terms of temperature-dependent dispersion parameters. However, it is well known from Monte Carlo simulations [4] that energetic disorder often gives rise to a power-law decay, even when the transport is not dispersive. The distinction between dispersive and nondispersive transport can only be made through the scaling with *L*. When the transient “mobility” scales with *L*, like in Eq. (2), the transport is dispersive. For nondispersive transport, the mobility is a genuine material parameter, independent of *L*. Although the scaling with *L* is not discussed by Lebedev *et al.* [9], they observed a transient mobility that agrees well with the DC mobility, which indicates nondispersive transport.

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In the following, we demonstrate that the differences between our results, based on delayed EL, and the results in Ref. [9], based on TOF, can be attributed to the different length scales used. By increasing the thickness of the PPV layer up to 820 nm, which is in the range used for TOF [9], we observe a transition from dispersive to nondispersive transport, in agreement with theoretical predictions.

2. Results and Discussion

In Fig. 1, the transient hole mobility, as calculated from the observed delay times τ_d according to Eq. (2), is shown as a function of the electric field E for different layer thicknesses at room temperature. The τ_d have been determined using the delayed EL technique described in Ref. [5]. While the mobilities in the samples with thickness up to 460 nm clearly follow the scaling law [Eq. (2)] as a function of E and L , the mobility in the 820 nm sample is much lower and shows a weaker field dependence. Instead, this mobility agrees well with the DC mobility [Eq. (1)], which strongly suggests a transition towards nondispersive transport. Furthermore, as shown in Fig. 2, the temperature dependence of the mobility in the 820 nm sample is also in reasonable agreement with the DC mobility.

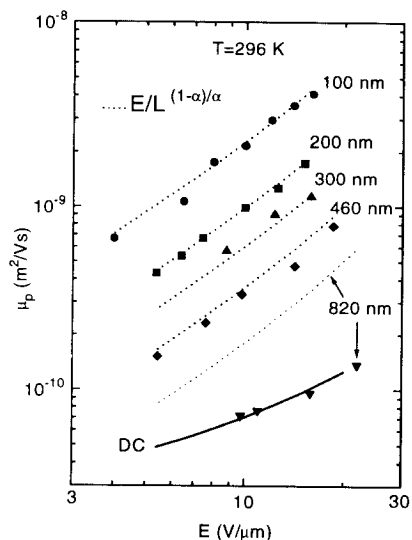


Fig. 1. Hole mobility μ_p , as calculated from the observed response times using Eq. (2), as a function of the applied electric field E (symbols) for LEDs with various L . The dotted lines denote the hole “mobilities” as expected from continuous-time random walk theory; the solid line denotes the DC hole mobility as obtained from J - V measurements.

Let us now compare this result with a theoretical estimate for the dispersive-nondispersive transport transition. Monte Carlo simulations [10] as well as analytic theory [11] predict that dispersive transport in a structurally disordered material can only be observed on timescales shorter than a certain demarcation time $t_0 = v^{-1} \exp(1.73 \gamma n^{-1/3})$. Here, v is an attempt rate, γ is the wavefunction overlap parameter, and n is the site density. After this time, the carrier has surveyed a representative fraction of the sample sites and the transport becomes nondispersive. Using a phonon frequency $v_0 = 10^{13}$ Hz and the average activation energy

to $\Delta_0 = 0.34$ eV, the attempt frequency $v = v_0 \exp(-\Delta_0/k_B T) \approx 10^7$ Hz at room temperature. Substituting this result, together with the relation $\alpha = 3 \gamma^{-1} n^{1/3}$ (Ref. [11]) and $\alpha = 0.45$ into the expression for the demarcation time, we find that t_0 lies in the millisecond regime, which is in fair agreement with the observed transit times for $L = 820$ nm.

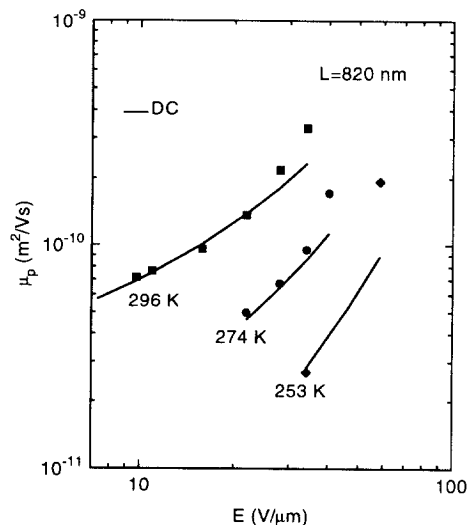


Fig. 2. Hole mobility μ_p , as calculated from the observed response times using Eq. (2), as a function of the applied electric field E (symbols) for a LED with $L = 820$ nm at different temperatures. The solid lines denote the DC hole mobility at these temperatures as obtained from J - V measurements.

3. Conclusion

The transit time of holes in PPV based LEDs is governed by structural disorder-induced dispersive transport. Upon increasing the PPV layer thickness a transition to nondispersive transport is observed. In this regime, the hole transit time corresponds well with the DC hole mobility found in stationary experiments [2]. Since time-of-flight experiments on PPV require layer thicknesses ≥ 700 nm [9], structural disorder-induced dispersive transport is not likely to be observed with this technique.

4. References

- [1] J.H. Burroughes *et al.*, Nature (London) 347 (1990) 539.
- [2] P.W.M. Blom, M.J.M. de Jong, and M.G. van Munster, Phys. Rev. B 55 (1997) R656.
- [3] P.M. Borsenberger and D.S. Weiss, Organic Photoreceptors for Imaging Systems, Dekker, New York, 1993.
- [4] H. Bässler, Phys. Status Solidi B 175 (1993) 15.
- [5] P.W.M. Blom and M.C.J.M. Vissenberg, Phys. Rev. Lett. 80 (1998) 3819.
- [6] H. Vestweber *et al.*, Synth. Met. 64 (1994) 141.
- [7] S. Karg *et al.*, Synth. Met. 67 (1994) 165.
- [8] H. Scher and E.W. Montroll, Phys. Rev. B 15 (1977) 2062.
- [9] E. Lebedev *et al.*, Appl. Phys. Lett. 71 (1997) 2686.
- [10] B. Ries and H. Bässler, Phys. Rev. B 35 (1987) 2295.
- [11] B. Movaghar *et al.*, J. Stat. Phys. 30 (1983) 315.