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Frequency dependent carrier mobility in polymer LED's

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Abstract

The charge mobility in polymer LED's depends on the polymer and its packing. In the latter various length scales play a role. To study this aspect, we performed dielectric spectroscopy as function of electric field and temperature on OC_1C_{10}-poly(p-phenylene vinylene). At low frequencies the mobility (μ) is mainly determined by the energetic disorder of the weakest link. Around 100 GHz μ has increased by 6 orders of magnitude and the field dependence has changed significantly. At these high frequencies, which in this case are representative for the pair limit, the field dependence shows that still appreciable energy barriers have to be overcome. Together these data allow a detailed characterization of the charge transport in the LED.

Key words: Transport measurements, amorphous thin films, Poly(phenylene vinelyne) and derivatives, light sources

1. Introduction

For about a decade conjugated polymers, especially PPV derivatives are known to generate light due to recombination of electrons and holes injected via special electrodes [1]. It has also become clear that in polymer LED's holes have a much higher mobility than electrons and that the current is bulk space charge limited [2] implying that the current flow is directly governed by the charge-carrier mobility. Current - voltage experiments showed that by chemically modifying the PPV material the mobility can be changed by several orders of magnitude [3,4]. In all cases the DC-mobility is well described by the empirical $\ln \mu \propto \sqrt{E}$ law. The specific E and T dependence of the data could be fitted within the (correlated) Gaussian disorder model [5,6]. Although DC data represent an average (macroscopic) property, the fitting parameters give values for microscopic parameters, like the localization length, the distance between hopping centers and the amount of energetic disorder. Indeed, the values give a consistent picture of the influence of the molecular structure of the polymer on the charge transport. The parameters show that broken conjugation, which dilutes the charge-transporting fraction of the material, naturally enhances the distance between the sites and therefore limits charge transport, while in the fully conjugated materials it is the amount of energetic disorder[4] that prevails.

Here we present mobility (μ) data at 0.1 and 0.2 THz as function of electric field E at room temperature obtained by means of an ABmm vector network analyzer[7]. As shown earlier for composites[8] and chemically doped polymers [9], such data sets are very sensitive for the local structure and when combined with the low frequency values, are very instructive. We confine the discussion to fully conjugated OC_1C_{10}-PPV.

2. Data and Discussion

Bässler [5] has argued that charge transport in disordered organic conductors proceeds by means of hopping in a Gaussian site-energy (ϵ) distribution $g(\epsilon) = (2\pi\sigma^2)^{-1/2} \exp[-\epsilon^2/(2\sigma^2)]$, with σ the

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width of the distribution. As suggested by Monte Carlo simulations, in a correlated Gaussian disorder model such a distribution might lead to the following functional dependence of μ (with $\beta = 1/k_BT$) $\mu = \mu_\infty \exp\{-(3\beta\sigma/5)^2 + 0.78[(\beta\sigma)^{3/2} - 2]\sqrt{eaE/\sigma}\}$[6]. This expression fits the E and T dependence of the DC-data very well. For fully conjugated PPV $\mu_\infty = 5.1 \times 10^{-9}\mathrm{m}^2/\mathrm{Vs}$[10], $\sigma = 112$ meV, equilibration energy $\sigma^2/k_BT = 0.5$ eV and the site spacing $a = 1.2 \pm 0.1$ nm.

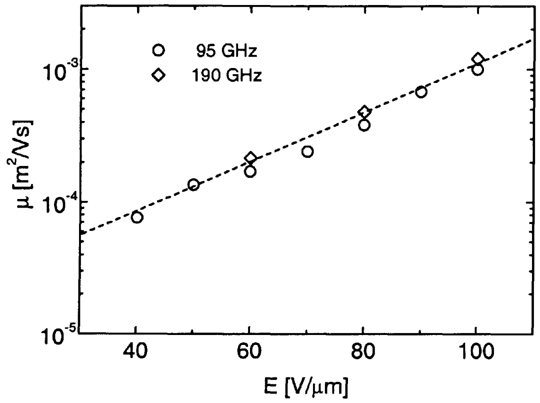

Fig. 1. Room temperature field dependence of μ of fully conjugated O_1C_{10}-PPV at 95 and 195 GHz. At these high frequencies the mobility is activated in E and hardly depends on frequency.

In Fig.1 we present the room temperature field dependence of μ for 95 and 190 GHz (data down to 9 GHz show a similar E dependence). The most striking difference of the high frequency with the DC data is the increase in the mobility by six orders of magnitude. While at room temperature the zero field mobility is $10^{-11}\mathrm{m}^2/\mathrm{Vs}$, the high frequency values are around $10^{-5}\mathrm{m}^2/\mathrm{Vs}$. Also the E-field influence is peculiar: in the DC experiments the mobility grows exponentially with $\sqrt{E}$, but at high frequencies μ increases almost exponentially with E (dashed line in the figure).

What causes these differences in the extrapolated zero-field mobilities? From the DC data μ_∞ was estimated as $5 \times 10^{-9}\mathrm{m}^2/\mathrm{Vs}$. The 10^3 times higher values at high frequencies clearly demonstrate the percolation character of the DC process, in which the weakest link dominates the mobility. The almost frequency independence of the mobility between 95 and 190 GHz is expected in the pair limit for sufficiently high frequencies. It means that the data give a picture of the hops between neighboring localized regions.

What can we learn from the dependence of μ on E at high frequencies, see Fig.1, and its qualitative difference with the low frequency data? In DC transport the mechanism behind the $\exp\sqrt{E}$-dependence is rather involved: in the percolation path many hops enter in series and parallel and all transitions probabilities will be changed by E to some degree. At high frequencies the transitions over the barriers are much more directly influenced by E and might be described by the expression of Pfifster[12,13], for which $\mu \propto (1/\beta eRE)\sinh(\beta eRE/2)$. This relation explains the observed field dependence and at room temperature gives a hopping distance R of a few nm, about the same value found for the hard hops from the DC data[4]. A similar distance was also derived from the analysis of the transients[13] and the frequency dependent electrical response of holes in this material[14]. The picture that arises is that in PPV hopping sites are separated by a distance of a few nm, even at short time scales, with barriers that are apparently (at least) of the order of 10^{-1} eV ($\sim$ the field energy of a 1 nm hop)[15].

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