

High intra-chain hole mobility on molecular wires of ladder type poly(p-phenylenes)

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ABSTRACT

The high frequency mobility of charge carriers on isolated ladder-type polymer chains with lengths ranging from 13 to 54 monomer units was measured. Experiments were performed on isolated chains in dilute solution and on bulk solid samples. The ac mobility of charge carriers measured at a microwave frequency near 30 GHz is strongly dependent on the chain length. The intra-chain motion of charge carriers can be described by one-dimensional diffusion between infinitely high reflecting barriers, representing the chain ends. Theoretical analysis of the experimental data yields an intra-chain mobility of charges on isolated ladder-type polymers in dilute solution near 600 cm²/Vs. For bulk solid samples the intra-chain mobility has a lower value of 30 cm²/Vs, which is attributed to energetic disorder in the bulk due to interactions between different polymer chains. With the high intra-chain mobility the ladder-type polymer is a promising candidate for future use as an interconnecting wire in molecular-sized electronics.

Keywords: ladder-type polymer, molecular electronics, intra-chain mobility

1. INTRODUCTION

The opto-electronic properties of conjugated polymers are of interest due to their applications in devices, such as organic light-emitting diodes, photovoltaic cells and field-effect transistors.¹⁻³ Using solution-processable polymers as semi-conducting compounds opens the way to flexible electronics and enables cheap and easy production of devices.^{4,5} Since the ongoing miniaturization of electronic components rapidly approaches the fundamental limit of photolithography, it is of interest to study the ultimate size limit for organic electronics. Single molecules are the smallest components that are likely to be functional in integrated circuits.⁶⁻⁸ In this context conjugated polymers have the prospect to be used as interconnecting molecular wires between molecular scale electronic components.

In all applications, the mobility of charge carriers is a key factor determining the performance of the electronic devices. Recently, charge carrier mobilities as high as 15 to 35 cm²/Vs have been found for organic oligoacene crystals.^{9,10} In these materials the calculated bandwidth is of the order of a few tenths of an electronvolt.^{11,12} For perfectly ordered conjugated polymer chains the bandwidth is of the order of a few electronvolt.¹³ As a consequence the mobility of charge carriers moving along ordered conjugated polymer chains is expected to be considerably higher than the mobilities found for oligoacene crystals; *i.e.* in the order of a few hundred cm²/Vs. The values found experimentally for the device mobility in conjugated polymers however, range from 10⁻⁷ to 10⁻¹ cm²/Vs, for derivatives of poly(para-

phenylenvinylene), poly(thienylenevinylene) and poly(thiophene),^{3,14,15} which is several orders of magnitude lower than expected.

This discrepancy is likely to be due to a combination of device-specific properties and intrinsic material properties. Contact effects are the most important of the device-specific properties, and determine the performance of organic optoelectronic devices to a large extent.^{16,17} Intrinsic properties of organic materials that limit the charge carrier mobility are mostly related to disorder in the material. Torsional disorder along the polymer backbone and conjugation breaks lead to barriers for charge transport.¹⁸⁻²⁰ Intra-chain disorder can also originate from inter-chain interactions. Places where the backbones of neighboring polymer chains closely interact might be energetically favorable and act as localization sites for charge carriers. Therefore, the potential energy landscape encountered by a charge moving along the backbone of a polymer chain can be significantly altered by the presence of neighboring polymer chains. In addition charge transport in a device can be limited by relatively slow inter-chain charge transfer steps and charge transport over grain boundaries (boundary junctions between different relatively ordered domains within the sample).

In this contribution it is shown that the intra-chain mobility for charge carriers moving along ordered ladder-type polymer chains can be considerably higher than the mobilities found for oligoacene crystals; *i.e.* in the order of a few hundred cm²/Vs. To obtain insight in the intra-chain motion of charge carriers and to circumvent device-specific effects and effects that result from inter-chain disorder, the mobility was determined by electrodeless time-resolved microwave conductivity (TRMC) measurements. The polymers studied, are the ribbon-like ladder-type poly(p-phenylenes) shown in the inset of Fig. 1.²¹ These polymers are structurally well defined with no torsional disorder, since the backbone is restricted to a planar geometry by a bridging carbon atom. Such a structure is optimal for charge transport because the electronic coupling between neighboring units is maximal for a planar structure.²² Comparison of the intra-chain mobility of isolated ladder-type polymer chains in dilute solution²³ with results for solid samples,²⁴ gives information about the effect of inter-chain interactions on the intra-chain charge transport properties.

2. RESULTS AND DISCUSSION

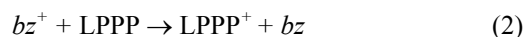
2.1 Intra-chain mobility along isolated polymer chains in dilute solution

To study the intra-chain mobility of charge carriers, the polymer chains were isolated from each other by dissolving them in benzene. The concentration in terms of monomers was equal to 0.315 mM. Charge carriers were generated by irradiation of the dilute polymer solution with a 10 ns pulse of 3 MeV electrons from a Van de Graaff electron accelerator. The high-energy electrons scatter on the solvent molecules and produce a close to uniform distribution of excess electrons and benzene cations with a known concentration. These excess electrons and benzene cations can diffuse towards the polymer chains, where they undergo charge transfer, thus yielding a charge on the polymer backbone.²⁵ The change in conductivity after the generation of charges was monitored by time resolved microwave conductivity (TRMC) measurements.²⁶ With this technique, the absorbance of microwave power as a result of the presence of (mobile) charges is monitored. With the TRMC method, it is possible to determine the high-frequency mobility of charges on isolated polymer chains in solution, without the use of electrodes.^{18,25,27,28}

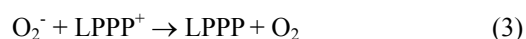
Fig. 1 shows the change in conductivity after the generation of charge carriers for the ladder-type polymers shown in the inset. In order to selectively study the mobility of holes along the polymer chains, the solution was saturated with oxygen. Due to the relatively high concentration (12 mM) and electron affinity of oxygen, the excess electrons generated during the electron pulse (e_{bz}^-) rapidly react with the oxygen molecules (O_2) forming the oxygen anion (O_2^-):



In this way, the transfer of negative charges to the polymer chains is prevented. Since the mobility of the oxygen anion in benzene is low (10⁻³ cm²/Vs)²⁹ as compared to the mobility of the positive charges along the polymer chain (see below), the contribution of the oxygen anion to the observed change in conductivity is negligible. The benzene cations generated during the 10 ns electron pulse (bz^+) react with the ladder-type polymer (LPPP) by a diffusion controlled reaction, yielding positively charged polymer chains (LPPP⁺)



As this reaction proceeds, an increase in the transient conductivity is observed on a timescale of hundreds of nanoseconds. This increase directly indicates that the positive charge on the polymer chain is more mobile than the benzene cation in benzene solution ($1.2 \times 10^{-3} \text{ cm}^2/\text{Vs}$).³⁰ On a timescale of ten to hundreds of microseconds a decrease in the conductivity signal is observed. The timescale of this decay decreases with increasing initial concentration of charge carriers, and can therefore be attributed to charge recombination between the oxygen anion and the positive charge on the polymer chain



A more extensive description of the reactions upon irradiation with high-energy electrons and the resulting transient conductivity of dilute polymer solutions can be found elsewhere.^{25,27}

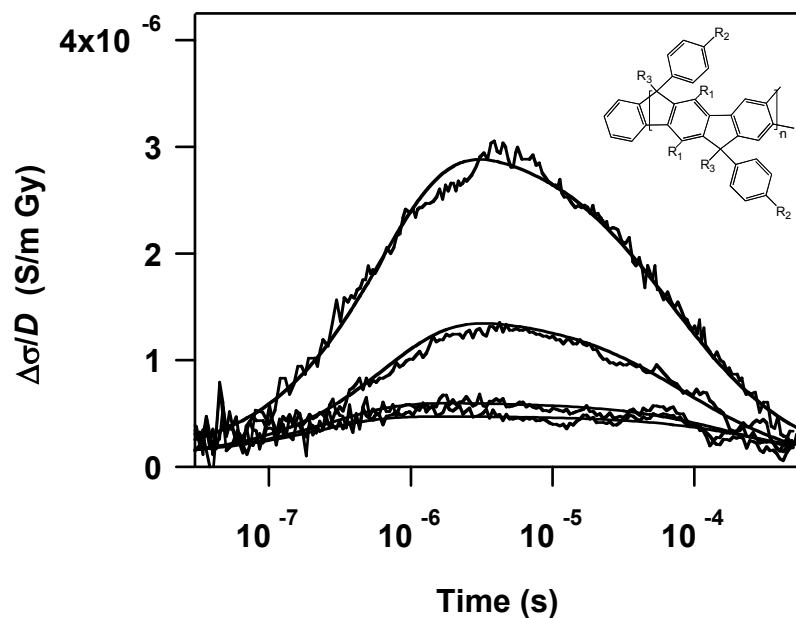


Figure 1. Conductivity due to positive charges on ladder-type polymers with a number averaged chain length of 13, 16, 35 and 54 repeat units, from bottom to top. The measurements were performed at a monomer concentration of 0.315 mM, a microwave frequency of 34 GHz, an electric field strength of 20 V/cm and an irradiation dose (D) of 21 Gy. The inset shows the chemical structure of the ladder-type polymers used in this study. $R_1 = n$ -hexyl, $\langle n \rangle = 13, 16, 35$; $R_2 = t$ -butyl, $R_3 = \text{phenyl}$, $\langle n \rangle = 54$; $R_2 = n$ -decyl, $R_3 = \text{C}$.

To obtain the high-frequency (ac) mobility for holes ($\mu_{ac, \text{hole}}$) from the measured conductivity shown in Fig. 1, the time dependent concentrations ($n_i(t)$) of charged species i must be known. These concentrations are obtained from a kinetic analysis of the conductivity transients, involving reactions (1)-(3).²⁵ The high-frequency mobility of holes on the polymer chains can now be obtained by using

$$\Delta\sigma(t) = e \sum_i \mu_{ac,i} n_i(t) \quad (4)$$

where e is the elementary charge and $\mu_{ac,i}$ denotes the high-frequency mobility of charged species i . The results for the conductivity as a function of time calculated according to Eq. (4) are shown in Fig. 1 by smooth curves.

As the average chain length increases, the high-frequency hole mobility is found to increase an order of magnitude from 0.025 via 0.036 and 0.10 to 0.24 cm²/Vs for an average chain length of 13, 16, 35 and 54 repeat units, respectively. This increase in mobility with chain length is indicative of hindrance of charge carrier motion by the chain ends, at least for chains as long as 35 monomers. The observation of this chain length dependence for the high-frequency mobility implies that the charge must diffuse over the entire length of the polymer chain, and encounter a chain end on a timescale in the order of one period of the oscillating electric field. For one-dimensional diffusion, the mean squared displacement as a function of time is given by $\langle x^2(t) \rangle = 2Dt$, where the diffusion constant D is related to the intra-chain mobility of the particle performing diffusive motion (μ_{intra}) and temperature (T) according to the Einstein relation $D = \mu_{intra} k_B T / e$. Using a displacement equal to a chain length of 35 monomers (30 nm), and a migration time equal to one period of the microwave field (30 ps), a value of 5 cm²/Vs can be deduced as a lower limit to the intra-chain mobility for positive charges moving along ladder-type polymers.

The high-frequency mobility of charges along polymer chains can be described by considering the diffusive motion of the charge along a one-dimensional flat energy surface between infinitely high reflecting barriers, which correspond to the polymer chain ends^{23,24}. Comparison of the experimental results for different average polymer chain lengths with the mobilities calculated using this model gives the absolute value of the intra-chain mobility.

In the one-dimensional diffusion model the high-frequency mobility of the charge carriers (μ_{ac}) depends on the (radial) frequency (ω), the length of the polymer chain (a) and the intra-chain mobility (μ_{intra}). The high frequency mobility of charges can be expressed in terms of the intra-chain diffusion coefficient (D) according to^{23,24}

$$\mu_{ac} = 8D \frac{e}{k_B T} \sum_{k=0}^{\infty} \frac{[c_k]^2}{\left[\frac{D}{L^2 \omega} \right]^2 [c_k]^4 + 1} \quad (5)$$

where

$$c_k = 2\pi \left(k + \frac{1}{2} \right).$$

To obtain the high-frequency mobility as a function of the intra-chain mobility for the average polymer chain lengths used in the experiment, the high-frequency mobility calculated with Eq. (5) is averaged over the chain length distributions in the samples, which correspond to truncated Flory distributions with $5 \leq n \leq 75$.³¹ The length of the ladder-type chains (L) is the product of the number of repeat units and the length of one repeat unit (8.3 Å). The results for the four average chain lengths are shown in Fig. 2a.

For low intra-chain mobility, the high-frequency mobility is equal to the intra-chain mobility; the motion of charges is not significantly hindered by the chain ends during the oscillation period of the probing field. The high-frequency mobility starts to deviate from the intra-chain mobility when the intra-chain mobility exceeds a few tenths of cm²/Vs, since the charge carrier starts to encounter the ends of the polymer chain. As the intra-chain mobility increases to tens of cm²/Vs, scattering of the charge carriers at the chain ends starts to dominate the charge transport and the high-frequency mobility decreases with increasing intra-chain mobility.

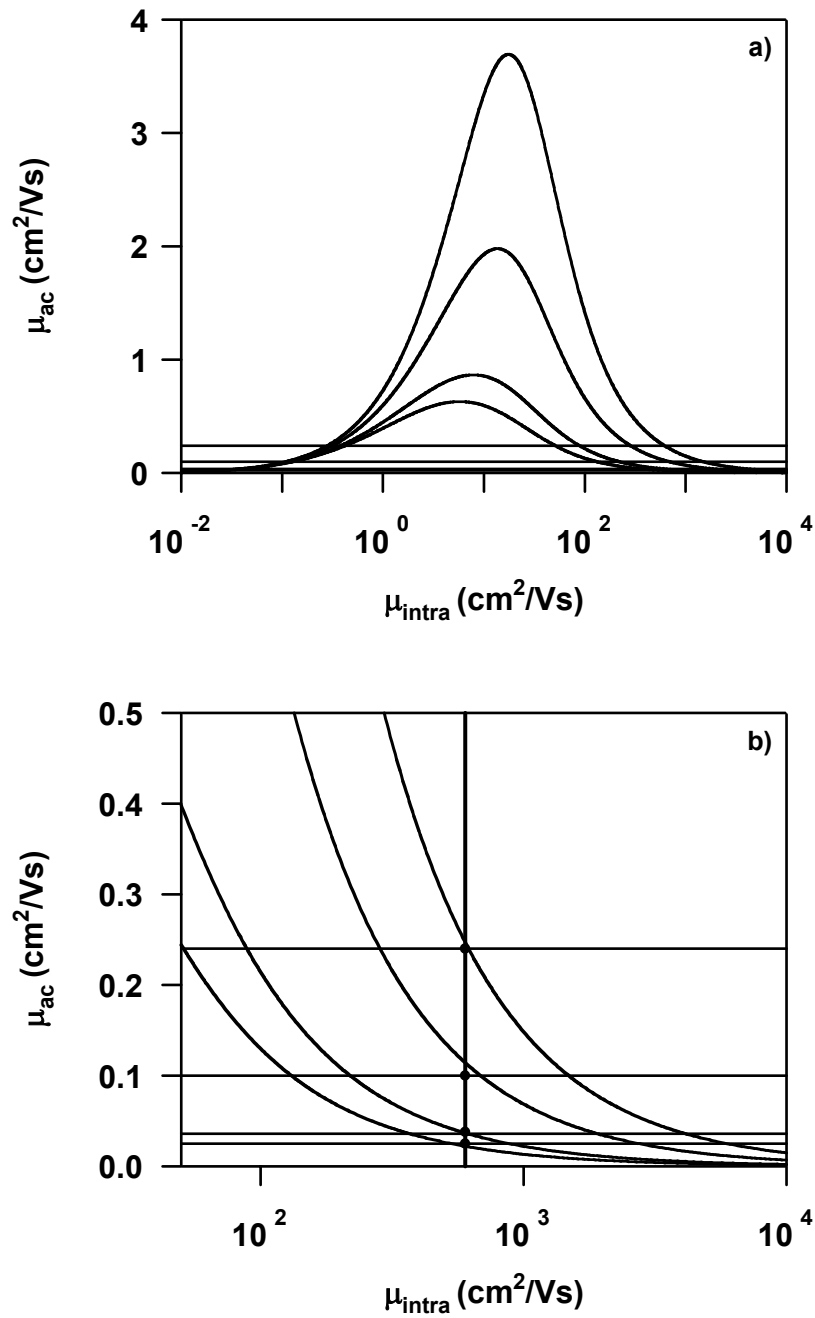


Figure 2. a) High-frequency mobility as a function of intra-chain mobility calculated with Eq. (5), for an average polymer chain length of 13, 16, 35 and 54 repeat units, from bottom to top. The horizontal lines represent the experimental data. b) The experimental values for the high-frequency mobility are reproduced by an intra-chain mobility close to 600 cm²/Vs.

To describe the experimental data in terms of the one-dimensional diffusion model discussed above, the intra-chain mobility that gives rise to the experimental values for the high-frequency hole mobilities for the four average chain lengths must be determined. Mobilities in the order of the experimental values, denoted by the horizontal lines in Fig. 2, are obtained both at a few tenths of cm^2/Vs and at a few hundred cm^2/Vs . At low intra-chain mobility, almost no chain length dependence would be observed for the high-frequency mobility probed by the microwave field, see Fig. 2a. Thus, the experimentally observed increase in the high-frequency mobility with chain length cannot be reproduced with an intra-chain mobility in the order of a few tenths of cm^2/Vs . This agrees with the estimate made above, that the intra-chain mobility must exceed $5 \text{ cm}^2/\text{Vs}$ in order to explain the chain length dependence of the experimental results. The data in Fig. 2b show that the measured high-frequency mobilities for different average chain length are reproduced by an intra-chain mobility close to $600 \text{ cm}^2/\text{Vs}$.

For an intra-chain mobility of $600 \text{ cm}^2/\text{Vs}$ the charge carrier motion along the polymer chains used in this study is severely hindered by the polymer chain ends. Hence, the one-dimensional diffusion model predicts a strong frequency dependence for the high-frequency mobility. Therefore, the high-frequency mobility was determined at a microwave frequency of 10.6 GHz for the ladder-type polymer in dilute solution. A high-frequency hole mobility of $0.032 \text{ cm}^2/\text{Vs}$ is obtained for the polymer with an average chain length of 54 repeat units. This value is significantly (7.5 times) lower than the mobility found for the same polymer at 34 GHz . The one-dimensional diffusion model in Eq. 5 gives a high-frequency mobility of $0.031 \text{ cm}^2/\text{Vs}$ at a microwave frequency of 10.6 GHz ($\langle n \rangle = 54$, $\mu_{\text{intra}} = 600 \text{ cm}^2/\text{Vs}$) which is in very good agreement with the experimental value. Thus the frequency dependence of the mobility substantiates the value of $600 \text{ cm}^2/\text{Vs}$ for the intra-chain mobility deduced from the chain length dependence.

The intra-chain mobility of $600 \text{ cm}^2/\text{Vs}$ is almost six orders of magnitude higher than found for the ladder-type polymer from time-of-flight (TOF) measurements³². The large difference demonstrates that intra-chain charge transport is much faster than motion of charges from one polymer chain to another. Relatively slow inter-chain charge transfer steps limit the TOF mobility.

2.2 Intra-chain mobility along polymer chains in bulk sample

In the experiments on solid samples approximately 3 mg of the ladder-type polymer in a poly(ethylene) container was placed in a microwave resonant cavity²⁴. The use of the resonant cavity makes it possible to determine both the real and imaginary components of the complex conductivity.²⁶ Charge carriers were generated by irradiation of the polymer with a pulse of 3 MeV electrons from the Van de Graaff electron accelerator. The change in complex conductivity due to the generation of charge carriers is given by

$$\Delta\sigma(\omega) = en[\mu_{h^+}(\omega) + \mu_{e^-}(\omega)] \quad (6)$$

where n is the number-density of electron hole pairs and $\mu_{h^+}(\omega)$ and $\mu_{e^-}(\omega)$ denote the complex mobility of holes and electrons, respectively. It should be noted that, in contrast to the experiments on dilute solutions described above, conductivity measurements on bulk solid samples yield information about the sum of the hole and electron mobilities, $\Sigma\mu = \mu_{h^+}(\omega) + \mu_{e^-}(\omega)$. The complex conductivity can be written as the sum of a real and an imaginary part: $\Delta\sigma(\omega) = \Delta\sigma_{re}(\omega) + i\Delta\sigma_{im}(\omega)$. The real part of the imaginary conductivity is due to motion of charges in phase with the probing electric field and can be detected as an increase in the absorption of microwave power in the polymer sample. The motion of charges out of phase with the probing electric field is determined by the imaginary part of the mobility. The imaginary part of the conductivity is related to the permittivity or dielectric constant of the medium ($\epsilon_0\epsilon$) and the radial frequency of the probing electric field according to $\sigma_{im}(\omega) = \omega\epsilon_0\epsilon(\omega)$. A change in imaginary conductivity upon irradiation with a 3 MeV electron pulse gives rise to a decrease in propagation velocity of the electromagnetic waves through the sample.

The change in real and imaginary conductivity after irradiation with a 3 MeV electron pulse are shown in Fig. 3 for the polymer with an average chain length of 35 repeat units. The imaginary part of the complex conductivity is larger than the real part of the complex conductivity for the ladder-type polymer. Any non-zero imaginary part of the complex

conductivity is indicative of hindrance of charge transport. Therefore, the results in Fig. 3 where the imaginary part exceeds the real part of the conductivity show that barriers to charge transport determine the charge transport properties in the ladder-type polymer to a large extent.

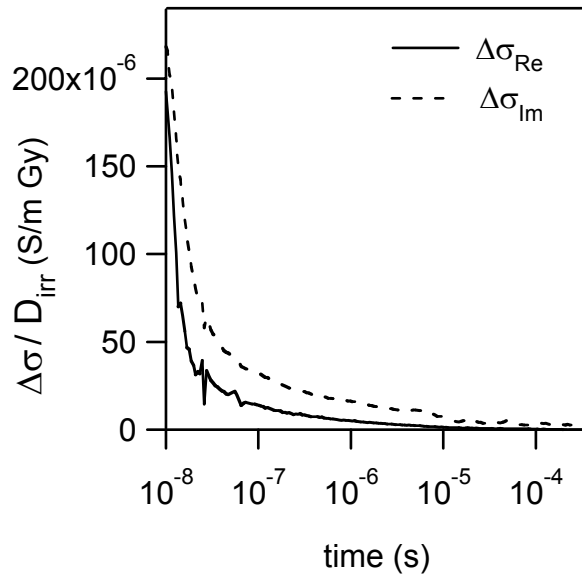


Figure 3. Real and imaginary dose-normalized conductivity after the creation of charges with a 3 MeV electron pulse for Phe-ladder polymer with an average chain length of 35 repeat units at an irradiation dose (D_{irr}) of 40 Gy.

The ratio of the imaginary and the real part of the complex mobility $\Sigma\mu_{im} / \Sigma\mu_{re}$ was found to be 2.2, 2.4, 3.8 and 4.0 for an average chain length of 54, 35, 16 and 13 repeat units, respectively. The chain-length dependence of this ratio directly shows that the out of phase motion of the charge carriers becomes more pronounced as the chain length decreases; *i.e.* charge carrier motion is more hindered on shorter chains. This is consistent with the idea that charges moving along a polymer backbone are hindered in their diffusive motion by the ends of the polymer chain.

A clear decrease in the ratio of the imaginary and real mobility is still observed as the average polymer chain-length increases from 16 to 35 repeat units. This shows that the charge must diffuse over the entire length of the polymer chain of (at least) 16 repeat units, and encounter a chain end on a timescale in the order of one oscillation period of the microwave field. A lower limit to the intra-chain mobility on ladder-type polymers in bulk solid can be found analogous to the discussion for isolated chains in dilute solution. For a displacement equal to a chain length of 16 repeat units (14 nm), and a migration time equal to one period of the microwave field (30 ps), a value of 1.3 cm²/Vs can be deduced as a lower limit to the intra-chain mobility in bulk solid.

In contrast to the experiments on dilute solutions, the charge carrier density at the end of the high-energy electron pulse is not known for solid samples, due to the unknown extent of charge recombination during the pulse. Hence, absolute values for the mobility cannot be directly obtained from the magnitude of the conductivity on the solid samples. However, the absolute values of the mobility can be obtained by theoretical analysis of the ratio of the imaginary and real mobility, using the theoretical model used to analyze the mobility data for polymers in dilute solution, as described in Section 2.1. The theoretical model describing the motion of charges along the polymer chain by one-dimensional diffusive motion between infinitely high reflecting barriers also provides an expression for the imaginary component of the mobility, analogous to the real component in Eq. (5). The value of the intra-chain mobility can be obtained by comparison of the calculated ratio of the imaginary and the real part of the complex high frequency mobility with the experimental results. In this way it was found that the experimentally determined ratios are reproduced by an intra-chain mobility close to 30 cm²/Vs.

The value for the intra-chain mobility of charges in solid samples is a factor 20 lower than the value of 600 cm²/Vs found for isolated chains of the ladder-type polymer in dilute solution. The difference in intra-chain mobility shows that the (polarization) interaction of a charge with the disordered environment due to neighboring polymer chains reduces the charge carrier mobility significantly. Inter-chain interactions cause a charge carrier to encounter a disordered energy landscape, which leads to a lower mobility.

3. CONCLUSIONS

The intra-chain mobility of charge carriers along ladder-type polymer chains was determined by theoretical analysis of the chain length dependence of the measured high frequency (ac) mobility near 30 GHz. The intra-chain dc mobility found in this way amounts to 30 cm²/Vs for bulk solid samples and 600 cm²/Vs for isolated polymer chains in dilute solution. The lower value for the bulk solid is due to disorder in the energetic landscape encountered by a charge carrier along a polymer chain, due to position dependent interactions of the charge with neighboring polymer chains. The intra-chain mobility for isolated ladder-type polymer chains in solution is more than three orders of magnitude higher than the highest literature values near 0.1 cm²/Vs, obtained from device measurements on conjugated polymers. This means that the limitation of the low mobility in conjugated polymers as compared with inorganic semiconductors can in principle be overcome by minimizing inter-chain transport. In the ultimate size limit for organic electronics, single molecules will be used both as electronic components and as interconnecting wires between these components. If these wires consist of individual ladder-type polymer chains the device performance will not be limited by the transport of charges along the chains. Therefore ladder-type polymers are promising candidates for use in molecular electronics.

4. ACKNOWLEDGMENTS

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