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Dimensionality of charge transport in organic field-effect transistors

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Application of a gate bias to an organic field-effect transistor leads to accumulation of charges in the organic semiconductor within a thin region near the gate dielectric. An important question is whether the charge transport in this region can be considered two-dimensional, or whether the possibility of charge motion in the third dimension, perpendicular to the accumulation layer, plays a crucial role. In order to answer this question we have performed Monte Carlo simulations of charge transport in organic field-effect transistor structures with varying thickness of the organic layer, taking into account all effects of energetic disorder and Coulomb interactions. We show that with increasing thickness of the semiconductor layer the source-drain current monotonically increases for weak disorder, whereas for strong disorder the current first increases and then decreases. Similarly, for a fixed layer thickness the mobility may either increase or decrease with increasing gate bias. We explain these results by the enhanced effect of state filling on the current for strong disorder, which competes with the effects of Coulomb interactions and charge motion in the third dimension. Our conclusion is that apart from the situation of a single monolayer, charge transport in an organic semiconductor layer should be considered three-dimensional, even at high gate bias.

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I. INTRODUCTION

In organic field-effect transistors (OFETs), the application of a gate bias voltage leads to the formation of a conducting channel in the organic semiconductor, commonly referred to as the accumulation layer, which extends from the source to the drain electrode. Several studies carried out on OFETs with π-conjugated molecules as organic semiconductors have suggested that charge transport in OFETs is confined to a thin layer close to the gate dielectric. Experiments on OFETs of sexithienyl with different coverage demonstrated that the first two monolayers sustain the whole source-drain current, where the second monolayer was suggested to provide percolation pathways for carriers present in both the first and the second layer.[1] In OFETs with dihexylquaterthienyl as the organic semiconductor the measured hole mobility exhibits maxima at coverages corresponding to one and two monolayers, and saturates at higher coverages.[2] More recently, an experimental study was performed by Shehu *et al*. that strongly indicates a non-two-dimensional nature of the charge transport in the channel of an OFET.[3] In their study, the authors monitored the source-drain current *in situ* and in real time during the deposition of pentacene on the gate dielectric. They suggested that the source-drain current in the transistor starts flowing when the first monolayer forms a percolating pathway and, depending on the deposition rate, saturates at a coverage corresponding to 2–7 monolayers. The authors also suggested that the number of active layers contributing to the current and the spatial distribution of the charge carriers is modulated by the growth method.

A continuum model assuming a semi-infinite and structureless semiconductor on top of a dielectric cannot explain the above experimental findings. According to such model, the screening depth of the gate electric field penetrating the organic semiconductor (the Debye length) is of the order of a nanometer,[4] which is of the same order as the intermolecular distance. Clearly, a proper description of the experimental situation should take into account the discrete nature of the charge-transport sites. A theoretical study of charge transport in OFETs in which hopping of charges between discrete transport sites was modeled by Monte Carlo simulations, taking into account energetic disorder and Coulomb interactions between the charges, showed that the charge carriers are confined in a conducting channel in the organic semiconductor with a thickness of about 5–6 nm, for realistic gate biases.[5] It was shown that most of the charges reside in the first monolayer and that the charge distribution decays rapidly with increasing distance into the organic semiconductor. The authors showed that with increasing gate bias the most significant increase of the charge-carrier density occurs in the first monolayer. This increased carrier density, together with the mobility-enhancing state-filling effect,[6–9] was used to explain the observed increase in mobility with increasing gate bias. The authors also claimed that despite the fact that the accumulation layer extends a few nanometers into the semiconductor, the gate electric field makes the charge transport essentially two-dimensional, implying that only the first, or maybe the first and the second, monolayer contribute to the charge transport.

In the present paper we will argue that the latter understanding of charge transport in OFETs is not entirely correct. We will show that hopping in the direction perpendicular to the semiconductor layer is an essential element of charge transport in the channel of an OFET, even for high gate biases, and that the transport is therefore non-two-dimensional.

Qualitatively, our arguments are based on the following. We first consider an OFET with a single monolayer of an organic semiconductor, as shown in Fig. 1(a). Energetic disorder in the semiconductor leads to a broadened density of state (DOS). The state-filling effect implies that with an increasing carrier density the carrier mobility increases.[6,7] In Ref. 6 an exponential DOS was considered and in Ref. 7 a Gaussian DOS. The latter is nowadays considered to be more realistic and this is therefore the DOS that we will consider in the present work. At low carrier density, the average distance between charge carriers is so large that they behave

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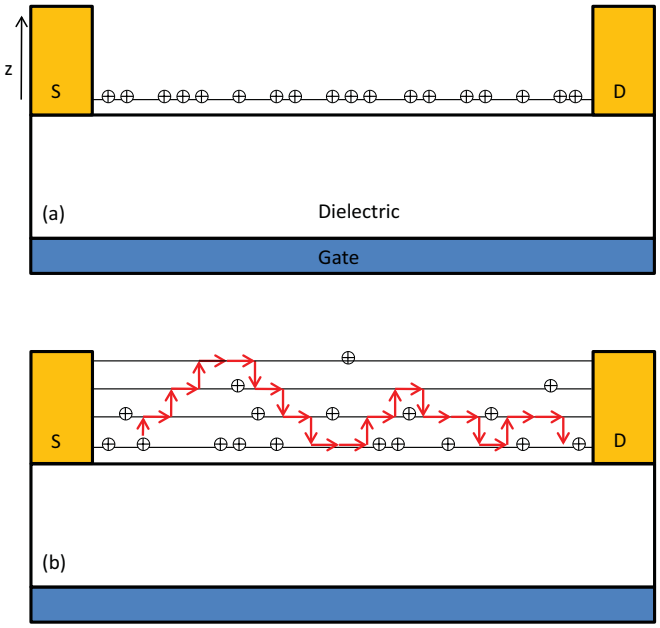

FIG. 1. (Color online) Schematic drawing of an OFET with (a) one and (b) four monolayers. S and D denote the source and drain electrodes, respectively. The monolayers are shown as horizontal lines. Circles with a plus sign represent charge carriers (holes). The total number of charge carriers is the same in (a) and (b). Red arrows in (b) indicate a possible trajectory a carrier may follow from source to drain.

independently from each other. The charge carriers occupy the low-lying states in the tail of the DOS and the energy barriers for charge transport are large, which results in a low mobility. Above a certain critical density,[8] the average energy of the charges increases substantially with increasing density, as the lowest-energy states are already filled. The activation energy for transport decreases, resulting in a higher mobility. In addition, there is the competing effect of Coulomb interactions between the charges. With increasing carrier density, also the Coulomb fields of the randomly distributed charges become more important. This leads to a broadening of the effective DOS and to a decrease in the mobility.[10] These two competing effects determine the carrier-density dependence of the mobility in an OFET consisting of a single monolayer.

Now consider the situation that additional monolayers of organic semiconductor are added on top of the first one, as shown in Fig. 1(b). With the same applied gate bias, the total number of charges will be the same. Some of the charges will move to the additional layers, so that the charge density in the first layer decreases. To understand charge transport in this situation, we have to account for the following effects:

(i) State-filling effect: Since the charges now occupy a larger volume, the average carrier density decreases. The decrease in carrier density has the tendency to decrease the carrier mobility.

(ii) Coulomb interactions: Since the charges now occupy a larger volume, the Coulomb interactions between the charges are reduced and this has the tendency to increase the carrier mobility.

(iii) Availability of new pathways: When other monolayers are added, many more pathways become available for a charge to move from source to drain electrode. This also has the tendency to increase the carrier mobility.

Whether the carrier mobility increases or decreases with the addition of monolayers depends on which of the above mentioned effects is dominant. If the state-filling effect dominates over the other two effects, the carrier mobility will decrease on addition of monolayers. On the other hand, if the joint effect of reduced Coulomb interactions and the availability of new pathways is dominant, the charge-carrier mobility will increase. One would furthermore expect that the effect of adding monolayers depends on the gate bias. For a higher gate bias the carriers will be pulled closer to the gate dielectric and one would therefore expect a weaker effect of increasing the thickness of the semiconductor layer. Following this line of thought, one might also expect that decreasing the gate bias can have an effect similar to that of adding additional monolayers in that it enables charge carriers to move farther away from the interface with the gate dielectric. We will show in this paper that this can indeed be the case.

The magnitude of the state-filling effect depends on the strength of the energetic disorder, specifically on $\sigma/k_{\mathrm{B}}T$, where σ is the width (standard deviation) of the Gaussian energetic disorder in the organic semiconductor and $k_{\mathrm{B}}T$ the thermal energy.[7,8] Therefore, the outcome of the competition between the above three effects will depend on the value of $\sigma/k_{\mathrm{B}}T$. Even a nonmonotonic dependence of the mobility on the number of monolayers may result. In the present paper, we will investigate theses issues in depth. As in Ref. 5, we will make use of Monte Carlo simulations of charge transport, taking into account the energetic disorder and the Coulomb interactions between the charges.

The paper is built up as follows. In Sec. II, we present the theory and the methods used. We describe our Monte Carlo approach and discuss in detail our method to take into account Coulomb interactions in an OFET geometry. In Sec. III we present and discuss the results obtained from Monte Carlo simulations for different values of the energetic disorder, thickness of the semiconductor layer, and gate bias. Sec. IV contains a summary and the main conclusions.

II. THEORY AND METHODS

In this section we describe our Monte Carlo approach for calculating the current and the distribution of charge carriers in an OFET. We model the organic semiconductor as a set of N_z monolayers stacked on top of each other in the z direction to form a regular three-dimensional lattice. Each monolayer is modeled as a square lattice of sites with lattice constant $a = 1$ nm, a typical value for organic molecular semiconductors. The vertical distance between the monolayers is also taken as a. The simulation box then has $N_x N_y N_z$ lattice sites, where N_x and N_y are the number of sites in the x and y direction, respectively. Periodic boundary conditions are taken in the x and y directions.

We assume that the charge transport occurs by phonon-assisted hopping of charges between sites. We choose to model

this by Miller-Abrahams hopping rates[11]

$$W_{ij} = \nu_0 \exp[-2\alpha R_{ij} - \beta(E_j - E_i)], \quad E_j \geqslant E_i,$$
$$W_{ij} = \nu_0 \exp[-2\alpha R_{ij}], \quad E_j < E_i. \tag{1}$$

Here, $\beta = 1/k_B T$, ν_0 is a phonon frequency, $R_{ij} \equiv |\mathbf{R}_i - \mathbf{R}_j|$ is the distance between sites i and j, and E_i and E_j are the on-site energies of sites i and j. For the inverse localization length of the wave functions α we have taken $10/a$.[7] For this value, farther than nearest-neighbor hopping is suppressed. The precise value taken for a then only influences a prefactor in the calculated current and mobility. It would be more realistic to take Marcus hopping rates,[12] but this would introduce an additional parameter, the reorganization energy. Moreover, it has recently been shown that for reorganization energies that are not much larger than σ the temperature dependence of the mobility for Marcus hopping is virtually the same as for Miller-Abrahams hopping, while the carrier-density dependence is exactly the same.[13]

The on-site energies E_i contain a random contribution $E_{\text{rand},i}$ that we will draw from the Gaussian DOS:

$$g(E) = \frac{1}{\sqrt{2\pi}\sigma a^3} \exp\left[-\frac{E^2}{2\sigma^2}\right]. \tag{2}$$

We note that the combination of Miller-Abrahams hopping and Gaussian disorder has been very successful in describing charge transport in various organic semiconductors.[7,14]

Apart from this random contribution, the energy difference in Eq. (1) contains two other contributions:

(i) The Coulomb interaction energy with all other charges, including the gate charge.

(ii) An electrostatic contribution $-eFR_{ijx}$ due to an electric field F, taken in the x direction of the lattice (e is the unit charge), due to a source-drain bias.

The Coulomb interaction of a charge with all other charges is split into three contributions in the following way.[15] (i) First, we take into account a short-range contribution, in which the Coulomb interaction energy with the charges within a sphere of radius R_c is taken into account explicitly. (ii) Next, we add a layer-averaged contribution, in which the Coulomb interaction energy with the other charges as well as that due to the applied gate electric field is taken into account in a layer-averaged way. Because this contribution also takes into account the layer-averaged Coulomb energy of charges in the disk-shape parts of the layers within the sphere, a double counting occurs. (iii) Therefore, we subtract a contribution due to these disk-shape parts to correct for this double counting. With increasing R_c, Coulomb interactions are taken into account in an increasingly exact way, with full exactness for $R_c = \infty$. For a well-chosen finite value of R_c we can obtain a good compromise between accuracy and simulation speed. For the results of the simulations discussed in the next section, we have taken $R_c = 15a$, which is more than sufficient.

Since charge transport occurs in a confined geometry, the Coulomb interactions between the charges have to be adapted to this geometry. The dielectric constant of typical organic semiconductors is not very different from that of silicon oxide, the most often used gate dielectric in OFETs. For simplicity we therefore take equal relative dielectric constants $\epsilon_r = 3.9$, the value for silicon oxide, for gate dielectric and semiconductor. The surface of the gate dielectric is taken to be the $z = 0$ plane, with the z axis pointing into the organic semiconductor, as shown in Fig. 1(a). The dielectric contrast between the semiconductor and the ambient, assumed to have a relative dielectric constant of 1, cannot be neglected. Using an image-charge technique, the electric potential due to a charge e at (x_e, y_e, z_e) at an arbitrary point within the semiconductor (x, y, z) can be expressed as

$$\frac{1}{4\pi\epsilon_0}\left(\frac{e/\epsilon_r}{\sqrt{(x-x_e)^2+(y-y_e)^2+(z-z_e)^2}} + \frac{e_t}{\sqrt{(x-x_e)^2+(y-y_e)^2+(z-z_{e,\text{img}})^2}}\right), \tag{3}$$

where $z_{e,\text{img}} = [2(N_z - 1)a - z_e]$ is the z coordinate of the image charge in the ambient, ϵ_0 is the vacuum permittivity, and $e_t = e(\epsilon_r - 1)/\epsilon_r(\epsilon_r + 1)$ is the total bound charge induced by the charge e at the interface between the semiconductor and the ambient.

We performed Monte Carlo simulations for the charge transport in three-dimensional square lattices of $100 \times 100 \times N_z$ sites for different values of N_z. The thickness of the gate dielectric is taken to be the typical 200 nm, which is a typical value for OFETs. Other values of this thickness would simply imply a scaling of the gate voltage. For all the calculations we assumed a temperature of $T = 30\,^\circ$C. We start with an empty lattice and fill it with a prescribed number of charges that is determined by the applied gate bias. With our parameters a change of 1 V of the gate voltage corresponds to a change in surface charge density in the semiconductor layer of 10^{-3} nm^{-2}. We assume the charges to be holes, which means that a negative gate voltage is taken. After filling, hops of these charges are chosen with weights determined by the hopping rates Eq. (1). Hopping times are chosen from an exponential distribution with an inverse decay time equal to the sum of all possible hopping rates. An applied source-drain electric field of $F = 0.1\sigma/ea$ was taken in the simulations, which is well within the linear regime of the charge transport. After about 10 million simulation steps, a stationary situation is obtained and the current is determined. The results presented in the next section have been obtained by performing an average of the currents over 15 disorder configurations of the on-site energies, resulting in an error margin of at most 5% in the reported results.

III. RESULTS AND DISCUSSION

We start with analyzing the charge and current profile for different thicknesses of the semiconductor layer. In Fig. 2 we plot the fraction of the total charge and current in each monolayer for different layer thicknesses, for a relatively low, $V_G = -10$ V, and high, $V_G = -40$ V, applied gate bias voltage, and for weak, $\sigma/k_B T = 2$, and strong, $\sigma/k_B T = 5$, disorder. On increasing the thickness of the semiconductor layer, the fraction of charges and current in each monolayer decreases, as expected.

At a higher (more negative) gate bias the charge is pulled closer towards the gate dielectric; compare the upper with the lower curves in Figs. 2(a) and 2(b). The effect of

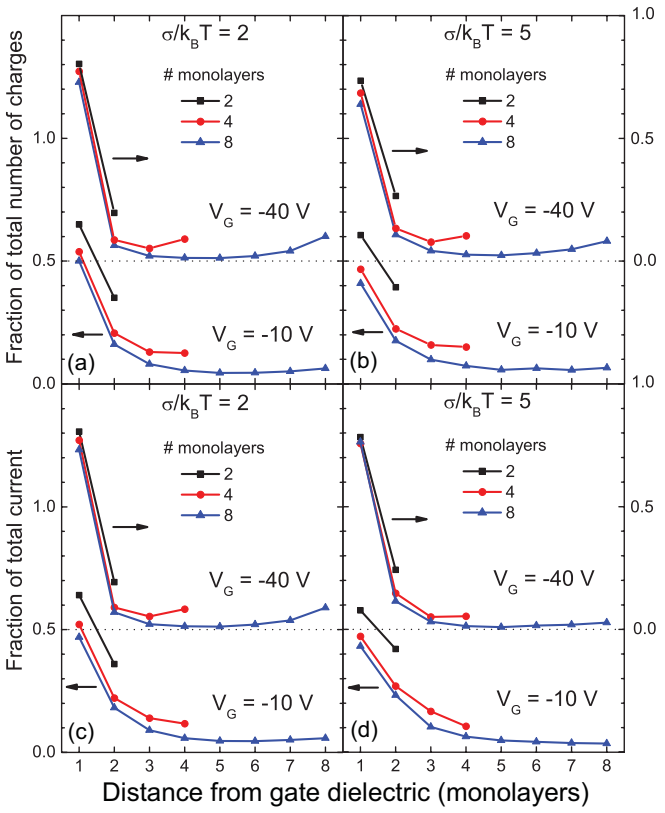

FIG. 2. (Color online) (a) and (b): Fraction of the total charge in each monolayer for different thicknesses of the semiconductor layer, expressed in the number of monolayers, and for two different values of the applied gate bias V_G. (c) and (d): Fraction of the total current in each monolayer. (a) and (c): Weak disorder. (b) and (d): Strong disorder.

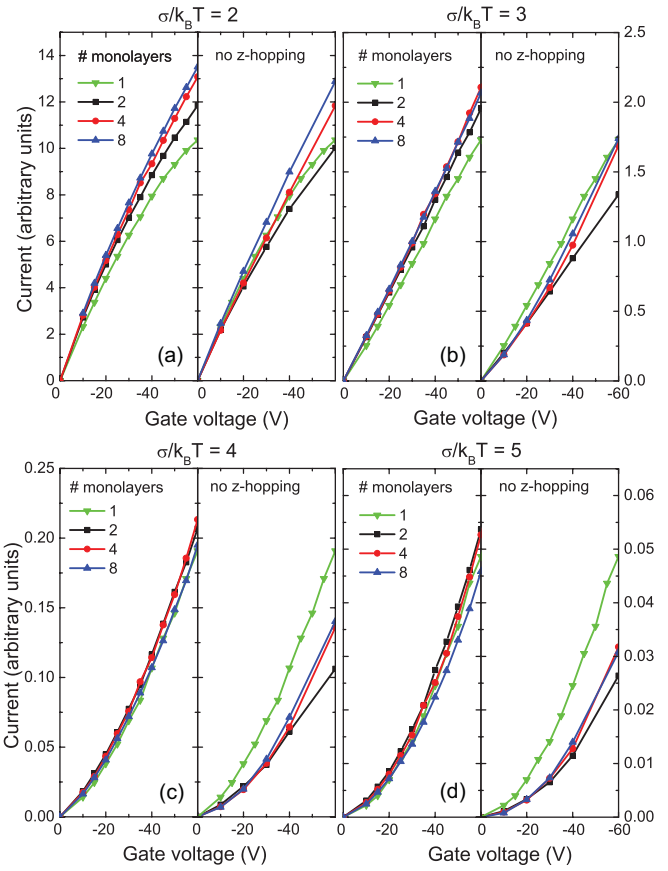

FIG. 3. (Color online) Total current as a function of gate bias voltage for different thicknesses of the semiconductor layer and for different disorder strengths. In the left panels hopping in all directions is allowed. In the right panels hopping in the z direction is switched off. Note the different vertical scales in (a)–(d).

increased disorder is a slight spreading of the charge over the semiconductor layer, but apart from that the effect of increased disorder is small. Somewhat surprisingly, the charge distribution does not decay all the way up to the last monolayer, but starts to rise again at the last monolayers. The reason for this upturn is the following. The charges in the first monolayers screen most of the applied gate electric field. The potential of the charges behind these first monolayers is not dominated anymore by the gate field, but by the Coulomb interactions between the charges. In order to minimize these interactions, charges move as far away from each other as possible. Since the charges are confined within the semiconductor layer, they accumulate near the boundary of the layer. The effect is largest for the highest gate voltage.

As expected from the charge profile the current flows mostly in the first monolayers; see Figs. 2(c) and 2(d). There is also an upturn in the current in the last monolayers, which is most clearly visible for weak disorder. For strong disorder the upturn is less visible. The reason for the latter is the strong carrier-density dependence of the mobility for strong disorder, which enhances the current in the first monolayers, where the carrier density is high.

In order to investigate the effects of state filling and dimensionality, we plot in Fig. 3 the current as a function of gate bias for different thicknesses of the semiconductor layer and different disorder strengths. The corresponding charge-carrier mobilities, which are proportional to the current divided by the gate bias, are plotted in Fig. 4. For weak disorder the mobility decreases as a function of gate bias, see Fig. 4(a), because at high gate bias the Coulomb interactions reduce the mobility of the carriers. However, for strong disorder the mobility increases with gate bias, see Figs. 4(c) and 4(d), because of the stronger state-filling effect than for weak disorder. In the intermediate case, occurring for $\sigma/k_BT = 3$ in Fig. 4(b), the effects of Coulomb interactions and state filling approximately cancel and a more or less bias-independent mobility arises, reflected by an approximately linear dependence of the current on gate bias in Fig. 3(b).

Richards and Sirringhaus investigated the charge-carrier mobility in an OFET of poly-dioctyl-fluorene-*co*-bithiophene (F8T2), before and after gate-bias stress.[16] The mobility was found to first sharply increase as a function of gate bias and then to either slightly decrease, for the OFET before stress, or increase, after stress. The initial increase could be due to non-Gaussianity of the DOS or by filling of traps. According to the above, the behavior at larger gate bias could indicate that the disorder strength is intermediate. After gate-bias stress, the disorder strength could very well be somewhat increased due to the Coulomb fields of trapped charges,[10] leading to an increasing instead of decreasing mobility with gate bias. In another paper the same authors showed that for

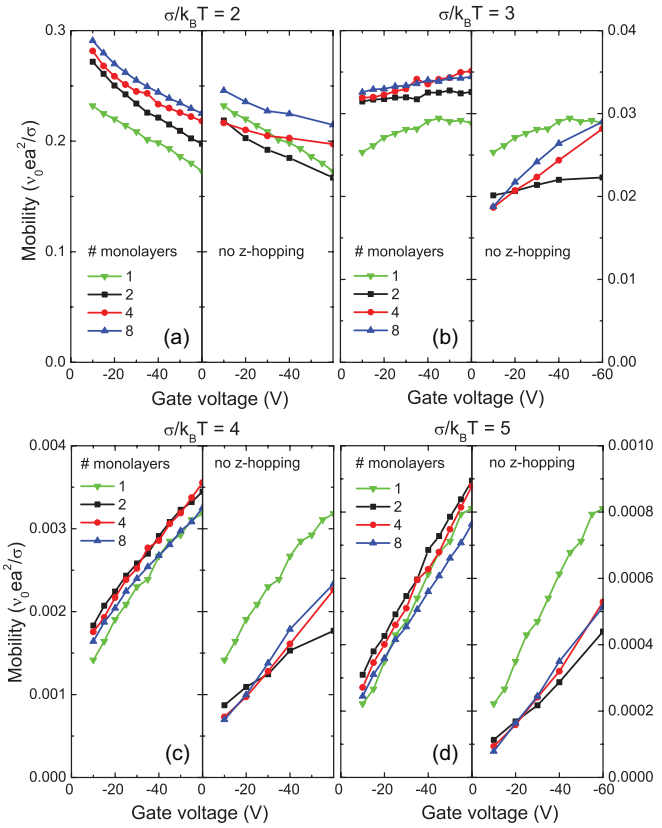

FIG. 4. (Color online) Charge-carrier mobilities as a function of gate bias voltage for different thicknesses of the semiconductor layer and for different disorder strengths, corresponding to the results for the current in Fig. 3.

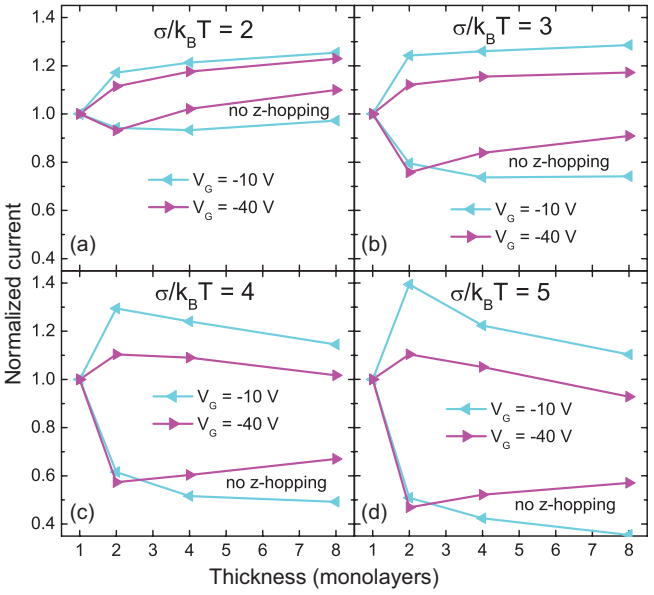

FIG. 5. (Color online) Current as a function of semiconductor layer thickness, normalized to the current for a single monolayer, for a low and a high gate voltage and for different disorder strengths. In the upper two curves in each graph hopping in all directions is allowed. In the lower two curves hopping in the z direction is switched off.

a poly(triarylamine) (PTAA) OFET the slope of the mobility versus gate bias curve depends on temperature.[17] After an initial steep rise at low gate bias, the mobility decreases with increasing gate bias at 300 K, whereas it continues to increase at 170 K. Again, such behavior is anticipated for intermediate disorder strength.

In the left panels of Figs. 3 and 4 hopping in all directions is allowed, whereas in the right panels vertical hopping in the z direction has been switched off (after the relaxation period). The latter reveals the effect of hopping in the third dimension. For weak disorder the reduction of the current by switching off vertical hopping is relatively modest. However, for strong disorder the reduction is about a factor of two. The reason for the reduction is that pathways as indicated in Fig. 1(b) are cut off. The effect is still important at high gate bias, when almost all charge is confined to the first monolayer. This demonstrates that transport in the organic semiconductor layer is essentially non-two-dimensional, even at high gate bias.

In Fig. 5 we plot the current at $V_G = -10$ V and -40 V as a function of the thickness of the semiconductor layer, for different disorder strengths. The current has been normalized to its value for a single monolayer. In the upper two curves in Figs. 5(a)–5(d) hopping in all directions is allowed, whereas for the lower two curves hopping in the z direction is switched off.

For weak disorder, see Figs. 5(a) and 5(b), the current for the case that hopping in all directions is allowed increases monotonically with increasing layer thickness. The reasons are the reduced Coulomb interactions and the larger number of available pathways for transport. The effect of the number of available pathways is clear from the difference with the current for the case that hopping in the z direction is switched off. This difference occurs in particular in the step from one monolayer to two monolayers.

For strong disorder a different behavior is observed: In the upper curves in Figs. 5(c) and 5(d) the current first increases when going from one monolayer to two monolayers and then decreases. The reasons for the initial increase are the same as for weak disorder: reduced Coulomb interactions and an increased number of available transport pathways. The latter effect is stronger than for weak disorder. This is clear from the results for the current without hopping in the z direction, which are lying considerably lower. When the thickness increases further, the large enhancement of the mobility due to charge confinement, present at strong disorder, gradually disappears. This leads to a decrease of the current for the case that hopping in all directions is allowed, which does not happen for weak disorder.

It is furthermore seen in Figs. 5(a)–5(d) that the change in the current by increasing the semiconductor layer is larger for high than for low gate bias, which is a result of the stronger confinement of the charge to the first monolayers; see Fig. 2. However, the effect of the gate bias rather strongly depends on the disorder. For weak disorder, the effect is smaller than for strong disorder; compare the upper two curves in Figs. 5(a) and 5(b) with those in Figs. 5(c) and 5(d). This difference is caused by the subtle disorder-dependent interplay between the effects of state filling, Coulomb interactions, and the availability of hopping pathways.

Interestingly, in the experiments of Muck *et al.* on OFETs with increasing coverage of dihexylquaterthienyl the satura-

tion value of the current with increasing coverage is lower than the value of the current at a coverage of one or two monolayers.[2] The authors tentatively attributed this to an increased contact resistance at high coverage, but they left open the possibility that the mobility itself decreases with increasing coverage. The decrease of the state-filling effect discussed in the present work provides a feasible explanation for a mobility decrease.

In the experiments of Shehu *et al*. on OFETs with increasing coverage of pentacene at low growth rates the current is highest at a coverage of two monolayers and then slightly decreases.[3] This behavior corresponds to the behavior reported here for moderate disorder [σ/k_BT between 3 and 4; see Figs. 5(b) and 5(c)]. For higher growth rates the current grows monotonically with coverage.[3] The absence of a maximum could in this case be explained by a less homogeneous growth, with substantial local height variations.

IV. SUMMARY AND CONCLUSIONS

We investigated the effects of dimensionality, disorder, and Coulomb interactions on charge transport in OFETs. The investigation is based on Monte Carlo simulations of hopping transport in the organic semiconductor, which is modeled as a regular array of sites consisting of a number of monolayers on top of a gate dielectric. We studied the influence of varying the thickness of the semiconductor layer, the strength of the disorder, and the gate bias.

We identified three effects that determine the dependence of the current on the semiconductor layer thickness: state filling, Coulomb interactions, and the availability of charge-transport pathways. The state-filling effect, which becomes more important with increasing disorder strength, has the tendency to increase the charge-carrier mobility with increasing charge-carrier density. On the other hand, Coulomb interactions between the charges have the tendency to decrease the mobility with increasing density. The availability of additional pathways for transport upon increasing the thickness of the semiconductor layer also has the tendency to increase the mobility. It is the competition among these three effects that determines whether the current increases or decreases with increasing semiconductor layer thickness.

We found that for weak disorder the current monotonically increases with increasing layer thickness. For this case, state filling has a minor effect. We attribute the increase of the current to the decrease of Coulomb interactions and to the increase in the number of available transport pathways. For strong disorder, we found that the current increases when going from one to two monolayers, but decreases when adding more monolayers. The initial increase of the current is explained in the same way as for weak disorder. The decrease of the current when adding more layers is attributed to the large mobility-enhancing effect of state filling for strong disorder. The state-filling effect decreases when the charge can spread over more monolayers and therefore the current decreases. We found that the effect of an increasing gate bias is comparable to a decreasing semiconductor layer thickness. Accordingly, the mobility increases with increasing bias voltage for high disorder strength, but shows a decrease for low disorder strength.

We confirm the result of previous modeling work on OFETs that the charge distribution is confined to the first one or two monolayers. However, by switching off hopping in the direction perpendicular to the semiconductor layer in the simulations, we concluded that hopping between monolayers is a crucial element of charge transport in OFETs, even at a high gate bias. Our final conclusion is therefore that charge transport in OFETs is essentially a three-dimensional process.

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