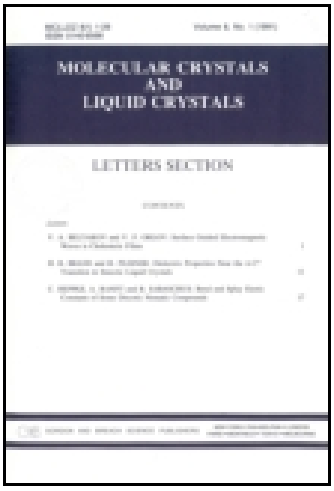

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Origin of Electric Field Dependence of the Charge Mobility and Spatial Energy Correlations in C60-Based Field Effect Transistors

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Origin of Electric Field Dependence of the Charge Mobility and Spatial Energy Correlations in C60-Based Field Effect Transistors

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We report on the influence of the lateral electric field on the charge mobility in organic field-effect transistors (OFET) based on C_{60} films with multigrain morphology. The experimental data were quantitatively described using a recent analytical model by accounting for the strong local electric fields in a multigrain transistor channel and for the energy correlation effects. To rationalize the presence of a correlated disorder in a non-polar C_{60} material, we show that randomly oriented permanent dipoles in organic gate dielectric layers can generate a significant dipolar disorder in an adjacent nonpolar semiconductor layer.

1. Introduction

Organic semiconductors offer a huge potential for the emerging flexible large-area electronics because they allow for a low cost device fabrication and a low-temperature processing of semiconductor layers compatible with flexible plastic substrates [1, 2]. Charge carrier transport is an important factor having a profound effect on ultimate device performance, notably, in an organic field-effect transistor (OFET). Therefore, it is of importance to improve the understanding of the conceptual premises of the electrical transport mechanisms in realistic

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organic electronic devices. The dependence of the charge-carrier mobility on electric field $\mu(F)$, is of particular interest as it bears on the fundamental nature of charge transport in organic semiconductors. In contrast to perfect organic crystals, where the charge-carrier mobility is normally independent of an electric field at room temperature [3], the charge-carrier mobility (μ) in disordered organic semiconductors typically increases with electric field in a Poole-Frenkel (PF) fashion, $\ln \mu \propto F^{1/2}$ [4, 5]. The latter is a consequence of an inherent energetic disorder and a thermally-activated hopping charge transport through the manifold of localized sites, which is commonly described by a Gaussian density-of-states (DOS) distribution of energetic width σ. The applied electric field tilts the DOS and thus lowers the average barrier height for energetic uphill jumps in the field direction [4].

The initial Gaussian disorder model suggested by Bässler and co-workers [4, 6] under the *premise of very low carrier concentration* was able to describe $\ln \mu \propto F^{1/2}$ dependence in terms of random (uncorrelated) energetic disorder, yet for a rather limited interval of electric fields [4, 7], which was often found to be too narrow to explain experimental results. To solve this discrepancy, an amended version of the formalism was developed by accounting for the spatial correlation of the energies of transport sites [8, 9]. The long-range energy correlations in organic disordered solids imply slowly varying static spatial fluctuation in the potential energy landscape and they conventionally arise due to interaction of charge-carriers with permanent dipoles [9] and quadrupoles [10] of adjacent sites. The accounting for the energy correlations was proven to be crucial to describe the Poole-Frenkel-type field dependence, at least in the *low-carrier-concentration* limit. Electric field dependence of the charge-carrier mobility is expected also in the *high-carrier-concentration* transport regime, e.g. for the OFET mobility, however, the reports of such a behavior in literature are rare [11–13]. Numerical simulations of the charge-carrier mobility in an energetically uncorrelated hopping transport system by Pasveer, Coehoorn, at al. [14, 15], based on the extended Gaussian disorder model (EGDM), have demonstrated a similar strong increase of the charge-carrier mobility with increasing both carrier concentration and electric field. Nonetheless, it has been a widespread opinion that at room temperature it is mainly the dependence on carrier concentration that plays an important role, whereas the electric field dependence becomes important at low temperatures and high electric fields [14]. One reason for such a notion is that the lateral electric field averaged over the transistor channel length is normally very low in OFET devices due to relatively long channels in OFETs, therefore more attention was paid to the mobility in sandwich-type diodes with space-charge limited (SCL) current. The electric field in the latter devices is normally much larger than the lateral electric field in an OFET due to very different inter-electrode distances.

The established hopping transport models normally predict that $\mu(F)$ should saturate at fields $F \leq 10^4$V/cm in typical organic disordered semiconductors even when the correlated disorder is taken into account [16, 17]. Nevertheless, an unexpected field dependence of the charge-carrier mobility in an organic field-effect transistor at low electric fields has been observed [18]. To rationalize the finding, it was suggested that the OFET mobility in multiple-grain channels can be controlled not by the lateral field averaged over the transistor channel (as conventionally assumed) but rather by the much stronger effective local electric fields generated in such inhomogeneous media [18]. The *concept of strong local electric fields* has been proven experimentally for a polycrystalline silylethynyl-substituted pentacene by performing combined charge transport and scanning Kelvin probe microscopy (SKPM) studies on OFET devices based on films from the same organic semiconductor but with and without grain boundaries [18]. The SKPM measurements were done under device operation and have clearly revealed strong voltage drops exactly

at grain boundaries, meaning that the local electric field in these boundaries is high. At the same time the electric field was found to be homogenous along the transistor channel containing a single crystallite/grain without a grain boundary. This agrees with the notion that grain boundaries control the macroscopic charge-carrier mobility in polycrystalline organic semiconductors [29–31]. It was shown [18] that the lateral field dependence of the OFET mobility in such materials can be quantitatively described, provided that just the local fields are used in the EGDM calculations instead of the average one. Since the actual ratio between local field at the grain boundaries and the averaged field is not amenable to analytical treatment, one can use a phenomenological field magnification parameter $q >> 1$ as a fitting parameter. Evidently the employment of this parameter q just results in a renormalization of the electric field Fused in the calculations.

The energy correlations are also required to describe the PF-type $\ln \mu \propto F^{1/2}$ dependence for the OFET mobility. Using numerical simulations Bouhassoune et al. [17] and Novikov [19] suggested a so-called Extended Correlated Disorder model (ECDM) [17, 19] to reproduce the PF dependence of the charge-carrier mobility at large carrier concentrations and applied it to organic diodes with SCL current [17, 20, 21]. The electric field dependent hopping transport in random organic media has recently been addressed analytically by Fishchuk et al. [22] who formulated an Effective Medium approximation (EMA) theory to describe the temperature-dependent hopping charge carrier mobility at *arbitrary electric fields* in the large carrier density regime taking into account the energy correlation effects. This model is an extended version of the previous EMA formalism [23, 24] within the ECDM approach and based on Miller-Abrahams jump rate and the effective transport energy concept. At large carrier concentration, typically realized in conductive channel of an OFET, a sizeable fraction of DOS is occupied and the charge transport is controlled by carrier jumps from states around the Fermi level to the transport energy level. In this transport regime the charge mobility depends also on the carrier concentration. The essential material parameters used in the model are the variance σ of the Gaussian DOS, the ratio of densities of occupied and total localized states n/N, and the ratio a/b of the intersite distance (a) and the localization radius (b) of the charged site. This theory yields the $\ln \mu \propto F^{1/2}$ dependence and it reproduces a compensation behavior for the temperature-dependent charge-carrier mobility upon *changing the electric field* provided that the carrier concentration is large enough [22]. This is reminiscent of the prediction of the empirical Gill relation [25]

$$\mu(F,T) = \mu_0 \exp\left(-\frac{E_a - \beta\sqrt{F}}{k_B T_{eff}}\right), \quad \frac{1}{T_{eff}} = \frac{1}{T} - \frac{1}{T^*}, \tag{1}$$

where E_a is the zero-field activation energy, β is a constant. The isokinetic temperature T^* is the so-called "Gill-temperature" at which extrapolations of the $\ln \mu vs 1/T$ lines intersect. It was shown [22] that the Gill-type dependence can be used at large carrier densities, as realized in conductive channels of OFETs, while it is not applicable when carrier concentrations are very low, e.g. for mobility data obtained in time-of flight (ToF) experiments. The EMA theory predicts that the Gill temperature (T^*) is not constant but should depend on carrier concentration n/N. This was shown to be a consequence of spatial energy correlations in organic disordered materials.

In the present study, we apply the recently suggested EMA model [22] to describe the electric field dependent electron mobility measured in OFETs based on C_{60} semiconductor films with multigrain morphology. The OFET mobility in C_{60} films features a Gill-type behavior regarding $\mu(T)$dependences upon varying the electric field and the Gill temperature

T^* was found to depend on carrier concentration in the transistor channel. We show that experimental observations for polycrystalline C60 films can quantitatively be described in terms of the EMA theory by accounting for the concept of strong local electric fields in the multigrain transistor channel [18] and for the energy correlation effects [17, 19, 22]. To rationalize the presence of energy correlations in non-polar C60 material, we demonstrate that dipolar disorder induced by the underlying polar gate dialectic layer is significant over a typical thickness of the conductive channel of C60 semiconductor and suggest that this might result in correlated disorder.

2. Experimental Results

C_{60}-based OFET devices were fabricated using divinyltetramethyldisiloxane-bis(benzocyclobutane) (BCB) as gate dielectric on ITO/glass substrates and C_{60} thin films as organic semiconductor as described previously [26, 27]. OFET charge-carrier mobilities were measured from transfer characteristics in the linear regime ($V_g - V_{th} >> V_d$, where V_g and V_d are the gate and drain-source voltages, respectively, and V_{th} is a threshold voltage) under vacuum conditions. Details of the experimental measurements were described elsewhere [26, 27].

The OFET charge-carrier mobilities were first measured at different source-drain voltages V_d (different lateral electric fields) and temperatures, but for the same V_g, which means for the same effective carrier concentration. Figure 1 (symbols) shows OFET mobility in C_{60} films as a function of lateral electric field at different temperatures plotted in $\log_{10}(\mu) \propto \sqrt{F}$ representation. It is evident that the OFET mobility features a PF-type dependence on lateral electric field in the range of very low fields of the order of $\sim 10^3$ V/cm generated by the applied source-drain voltage V_d and averaged over the conductive channel (length 35 μm). Theoretical fit of the experimental data by the EMA model [22] accounting for the energy correlations and using $q = 256$ as fitting parameter to account for the strong local fields in the conductive channel [18], is shown by solid lines in Fig. 1. The width of the DOS, $\sigma = 0.07$ eV, used in the fitting procedure was estimated for this material earlier [27]. The intermolecular distance $a = 1.4$ nm was assumed to be the same as in C_{60} crystals and $a/b = 5$ was taken as a representative value conventionally used in charge-carrier transport simulations for small molecule organic materials [4].

A fit to experimental data within this model, but for uncorrelated disorder, is shown by dashed curves in Fig. 1. Both uncorrelated- and correlated- disorder approaches provided seemingly a comparatively good description of the experimental data, although only the latter approach provides a strict PF-type field dependence in the considered field range. This implies that the present field dependences of electron mobility alone do not allow to distinguish unambiguously whether or not spatial energy correlations are important in this material. Dotted curves in Fig. 1 were calculated assuming a homogenous lateral electric field equal to the field averaged over the channel length. i.e. at $q = 1$. As one can see, no field dependence of the mobility is expected within the range of such low electric fields. This justifies the critical role of strong local fields suggested before [18] in the field dependence of the OFET mobility in C60 films with multigrain morphology (a typical morphology a C60 film used in this study is shown by an AFM image in the inset to Fig. 2).

Figure 2 shows the temperature dependence of the charge carrier mobility measured in C_{60}-based OFETs at different lateral electric fields (symbols) [26] and the values calculated

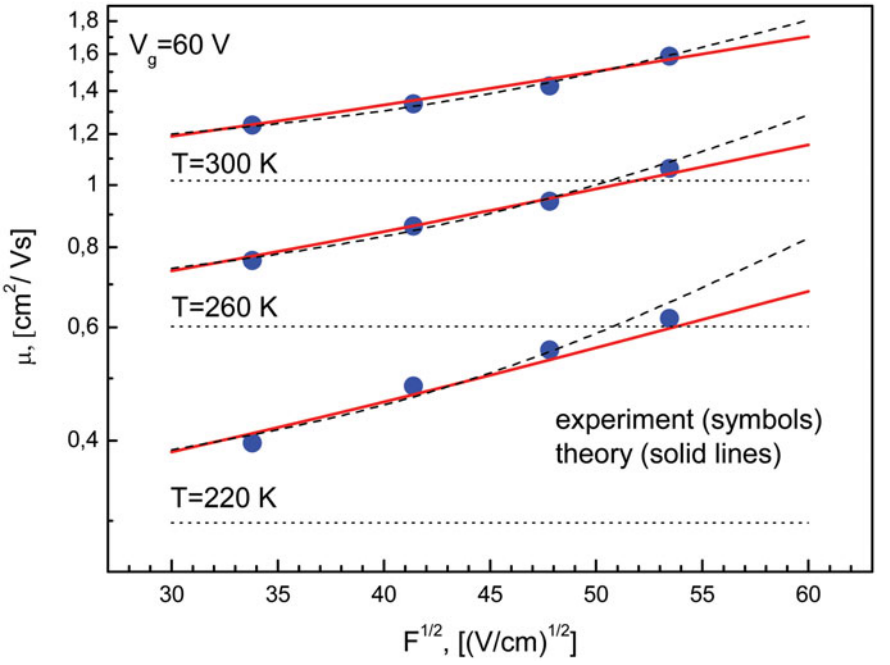

Figure 1. Experimental (symbols) lateral field dependence of the charge carrier mobility measured in C_{60}-based OFETs at different temperatures from Ref. [26] and theoretical fitting by the EMA model [22] (solid lines) for energy correlated disorder using the following set of parameters: $\sigma = 0.07$eV; $a/b = 5$; $a = 1.4$ nm; $n/N = 0.05$ and $q = 256$. Dotted curves were calculated for same correlated disorder system at $q = 1$assuming a homogenous lateral electric field. Dashed curves show a fitting of the experimental results with the same model but ignoring any energy correlations using $\sigma = 0.055$eV and $q = 100$.

by the EMA model [22] (bold curves) using the same set of parameters as in Fig. 1. The asymptotes (thin lines) to the calculated curves (bold curves) intersect at an finite temperature, thus yield the Gill-temperature $T^* \cong 429K$.

A remarkable observation in this study is that the Gill-energy $E_{Gill} = k_B T^*$, as determined from temperature dependences of the OFET mobility upon varying lateral electric field in the same device (c.f. Fig. 2), was found to depend on the applied gate voltage V_G (Fig. 3, symbols). This dependence has been predicted theoretically [22] and can also be successfully fitted by the present model using the same set of parameters as in Fig. 1; the calculated values are given by a solid curve in Fig. 3. The effects observed in Fig. 3 can only be fitted provided that energy correlated disorder is taken into account; E_{Gill} calculated for uncorrelated disorder is independent of V_G (dashed line in Fig. 3). It should be noted that a closely related effect has been observed before for the so-called Meyer-Neldel energy upon the applied lateral electric field [28]. The Meyer-Neldel energy, as determined from the temperature dependences of OFET mobility upon varying carrier concentration, was found to depend on the applied lateral source-drain electric field F_{SD} in C_{60}-based OFETs [28].

This suggests an important experimental test for the presence of correlated energy disorder in an organic semiconductor: Meyer-Neldel and Gill energy should manifest a dependence on lateral electric field and on carrier concentration, respectively. Besides, we

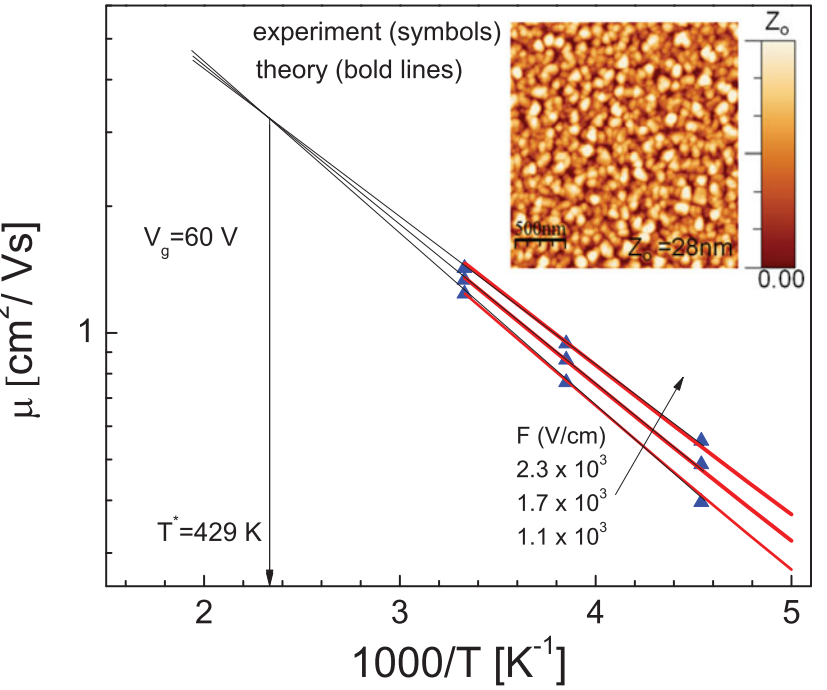

Figure 2. Temperature dependences of the OFET charge carrier mobility measured in C_{60}-based OFET at different lateral electric fields (symbols) [26] and theoretical fitting by the EMAC model (bold curves) using the same set of parameters as in Fig. 1. Inset shows a typical AFM image of a C_{60} film under investigation.

should point out some practical implications of the above result - as it was suggested in Ref. [22] from the experimentally determined dependence of E_{Gill} vs. F one can evaluate the energy disorder parameter σ and the local electric fields (the factor q).

3. Discussion

The field dependence of the OFET mobility observed in polycrystalline C60 films (Fig. 1) is basically similar to that observed before in pentacene-based OFETs [18] and suggests that the mobility in these films is controlled by grain boundaries. A prominent multi-grain morphology of the C60 films is demonstrated by the AFM image in Fig. 2. Thus, the recently suggested concept of strong local electric fields generated at grain boundaries has to be similarly invoked to describe the experimental data. We have recently proved this concept experimentally by performing combined charge transport- and SKPM studies on films from the same organic semiconductor but with and without grain boundaries [18]. The grain boundaries are known to limit charge-carrier transport in polycrystalline films by establishing major potential barriers between their more ordered domains [29–31]. In such a case upon applying a gate voltage to a channel with grain boundaries, the density of accumulated electrons within an individual grain is redistributed along the *external* lateral field direction. This creates an *internal* lateral electric field within the individual grain, which compensates the applied external field. This effect generates high local fields between the grains (i.e. at the grain boundary), while the field inside the grains

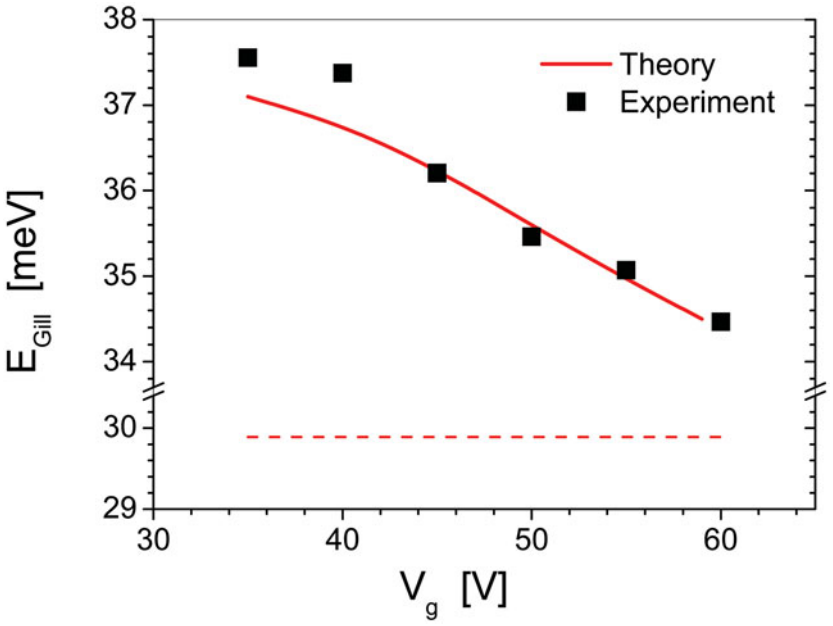

Figure 3. Experimentally determined Gill-energy $E_{Gill} = k_B T^*$ as a function of used gate voltage V_G. Fitting by the EMA model [22] with accounting for the energy correlations using the same set of parameters as in Fig. 1 is given by solid line. The dashed curve shows the theoretical dependence calculated upon ignoring any correlation effects.

is screened. It worth noting that the idea of large local fields has been already suggested long ago to describe charge carrier photogeneration yield in amorphous α-Si and Se [32, 33] and it was also proven experimentally. However, their origin was assumed to be due to potential fluctuations in disordered semiconductor and requires the presence of a sufficient concentration of equilibrium charges of opposite signs. Thus, this model is not applicable to organic materials where no considerable chemical doping occurs.

As mentioned above, the PF-type field dependence and the observed dependence of the Gill temperature T^* on carrier concentration (gate voltage) can be explained by the present EMA model only when the correlated energy disorder is present. It is commonly believed that the latter arises due to charge-dipole [9] or charge-quadrupole interactions [10]. Another origin can be inhomogenities in the electronic polarization energy [34], resulting from molecular density fluctuations in a material, due to microscopic regions that are under compression or dilation. The latter are well expected to occur in the regions around grain boundaries in such multigrain C60 films, therefore molecular density fluctuations should result in slowly varying potential fluctuations in them. Since C60 molecules have no permanent dipole or quadrupole moment, the intrinsic dipolar disorder is definitely impossible for a neat C60 film. However, one has to take into account that in OFET configuration the charge-carrier transport actually probes a very thin layer of the semiconductor - a few molecular layers directly at the interface with the organic gate dielectric which is a polar material. The organic dielectric BCB is formed by molecules bearing a permanent dipole moment. Below we demonstrate that this can generate a correlated dipolar disorder spreading into a nonpolar organic layer.

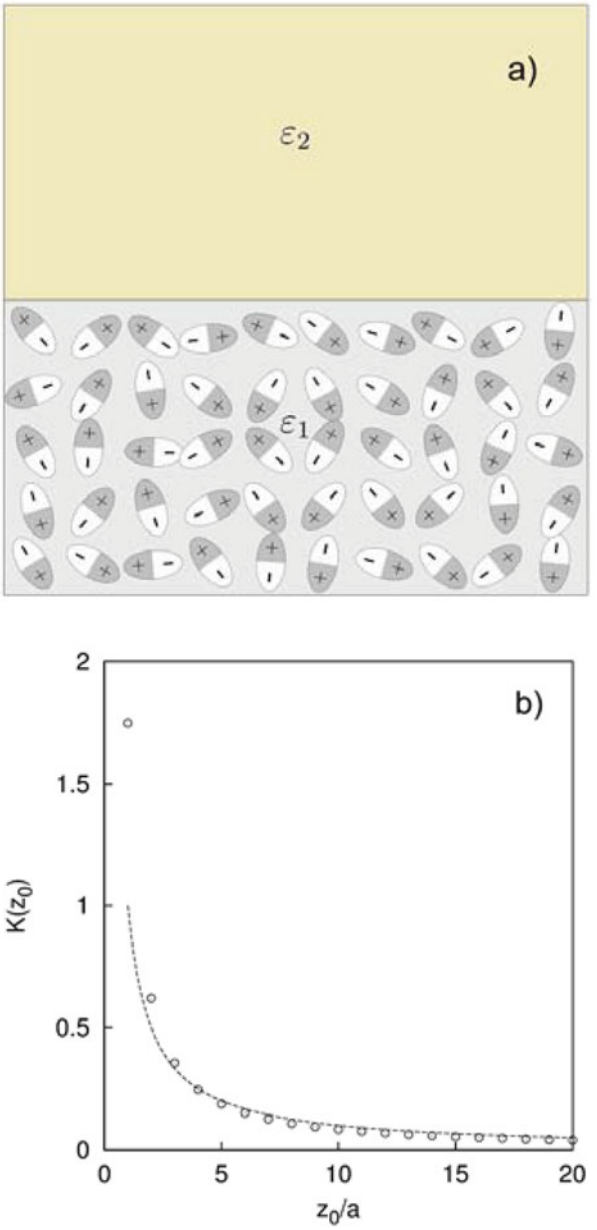

Figure 4. (a) Two organic layers separated by a plane interface; (b) $K(z_0)$factor calculated for the discrete lattice model by Eq. (3) (symbols). Dashed line shows the behavior of $K_c(z_0) = a/z_0$, where $K_c(z_0)$ is the corresponding continuous approximation according to Eq. (5) and assuming $a_c = a$.

Let us consider two organic layers separated by a sharp planar interface as schematically shown in Fig. 4a, with dielectric constants ε_1 and ε_2 for the lower and upper layers, respectively. Randomly oriented permanent dipoles are immersed in the lower layer (we assume that dipoles are located at the sites of the simple cubic lattice with lattice spacing a). Let us calculate the variance $\sigma^2(z_0)$ of the dipolar energy disorder in the upper layer

assuming the continuous approximation (here z_0 is distance from the interface). We ignore here any other sources of the energy disorder in the upper layer and assume it could be adequately described by the only parameter, namely, the static dielectric constant ε_2.

A point dipole with the moment p located in the lower layer at the position $\vec{r}_0$ generates in the upper dielectric the potential [35]

$$\varphi(\vec{r}) = \frac{2}{\varepsilon_1 + \varepsilon_2} \frac{\vec{p}(\vec{r} - \vec{r}_0)}{|\vec{r} - \vec{r}_0|^3}. \tag{2}$$

This expression differs from the conventional simple expression for the potential in the uniform medium by the replacement of ε with $(\varepsilon_1 + \varepsilon_2)/2$. Hence, one can use the well-known expression [36] for the variance of the dipolar disorder in the uniform medium with the corresponding replacement of ε; the only difference is that we have to restrict the lattice summation to the half-space $z > 0$ (the interface is considered as XY plane and Z axis is directed to the bulk of the bottom layer)

$$\begin{aligned} \sigma^2(z_0) &= \frac{4e^2p^2c}{3(\varepsilon_1 + \varepsilon_2)^2} \sum_{z_n>0} \frac{1}{\left[x_n^2 + y_n^2 + (z_n + z_0)^2\right]^2} \\ &\approx \frac{8\pi e^2p^2c}{3(\varepsilon_1 + \varepsilon_2)^2 a^3} \int_0^\infty d\rho\rho \int_0^\infty dz \frac{1}{\left[\rho^2 + (z + z_0)^2\right]^2} = \frac{4\pi e^2p^2c}{3(\varepsilon_1 + \varepsilon_2)^2 a^3 z_0}, \end{aligned} \tag{3}$$

here c is the fraction of sites occupied by dipoles. In the continuous approximation the dipolar variance σ_b^2 in the bulk of the lower layer is given as

$$\sigma_b^2 = \frac{4\pi e^2p^2c}{3\varepsilon_1^2 a^3 a_c}, \tag{4}$$

where $a_c$is a short range cut-off included in order to avoid the contribution from the site where the carrier is located; typically, $a_c \cong a$.

Hence,

$$\frac{\sigma^2(z_0)}{\sigma_b^2} = \frac{\varepsilon_1^2}{(\varepsilon_1 + \varepsilon_2)^2} \frac{a_c}{z_0}. \tag{5}$$

In fact, the relation (5) is formally valid only at large distances from the interface, $z_0 >> a$. For the actual discrete lattice model (c.f. Eq. 3) the corresponding spatial factor

$$K(z_0) = \frac{\sigma^2(z_0)}{\sigma_b^2} \frac{(\varepsilon_1 + \varepsilon_2)^2}{\varepsilon_1^2} \tag{6}$$

is shown in Fig. 4b (open circles) and its continuous approximation by Eq. (5) is given by a dashed curve.

The correlation function $C(x_0, z_0)$ (here x_0 is the distance along the interface plane) could be calculated in a similar way, though the calculation is more tedious. We provide here only the most important result valid in the vicinity of the interface $x_0 >> z_0$

$$C(x_0, z_0) = 2\sigma_b^2 \frac{\varepsilon_1^2}{(\varepsilon_1 + \varepsilon_2)^2} \frac{f(s)}{x_0}, \quad s = \frac{z_0}{x_0}. \tag{7}$$

For $s << 1$ $f(s) \approx 1 - As$, and the constant $A \cong 1$. Hence, in the vicinity of the interface the correlation function of the induced disorder differs from the usual dipolar correlation function $C(\mathbf{r}) \propto \sigma^2/r$ [37] mostly by the numeric coefficient reflecting the decrease of σ^2 (in our case x_0 is equivalent to r).

The above calculations demonstrate that dipolar disorder induced by the underlying polar dielectric film is considerable for at least several C60 molecular layers next to the interface and it decays as $\propto 1/\sqrt{z}_0$ with the distance into the nonpolar film. Taking the dielectric constant for BCB and C_{60} as 2.7 and 4, respectively, one obtains that the dipolar disorder component in the first C_{60} molecular layer next to the interface, which plays the major role in the conductive channel, comprises $\sigma_{dip}^{C60} \approx 0.52\sigma_{dip}^{BCB}$ of the bulk dipolar disorder σ_{dip}^{BCB} of the BCB film, that is quite considerable. The dipolar disorder of the second C60 monolayer layer is $\approx 0.33\sigma_{dip}^{BCB}$. We should note that these results perfectly agree with recent force-field and quantum-chemical calculations [38] which demonstrated that the electrostatic interactions introduce a significant energetic disorder in the pentacene OFET-semiconductor layer in contact with the polymer chains bearing static dipoles.

Conclusions

The predictions of the EMA model for the electric field dependent charge-carrier transport are compared with experimental data obtained for the C_{60}-based OFETs. We show that the Gill- temperature can depend on carrier concentration and this is a consequence of spatial energy correlations in the material. We discuss the idea of strong local electric fields created in an OFET conductive channel formed in polydomain organic semiconducting films and show that this concept is crucial to explain the experimental observation of the lateral-field dependent OFET mobility also in C60 polycrystalline films. Fitting of the experimental data with the EMA theory allows estimating important material parameters as the width of the DOS distribution, characterizing the degree of energetic disorder. Finally we show that a dipolar gate dielectric layer can generate a significant dipolar disorder also in the adjacent nonpolar semiconductor over a length of typical thickness for the conductive channel, thus yielding the correlation function of the induced dipolar disorder in the vicinity of the interface that is quite similar to that for usual dipolar glasses. Thereby this can result in energy correlated dipolar disorder in channels made of an organic semiconductor with zero dipole and quadrupole moments.

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