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CURRENT FILAMENTATION AND DEGRADATION IN ELECTRONIC DEVICES BASED ON AMORPHOUS ORGANIC LAYERS

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Dedicated to the memory of Professor Vladimir Šips

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The recent advent of new flat-panel organic displays follows a long struggle for the extended device lifetime. Many modifications proposed along this way have been based on trial and error experimentation. In this paper, we show that some recent major improvements may have been implicitly related to fighting against the strong current filamentation, intrinsic for charge injection and conduction in organic amorphous thin films. We first recapitulate some major causes of current filamentation in thin amorphous organic layers. Then we consider the charge transport in devices with a high-mobility injection layer, with a smoothened organic heterojunction surface, that operate at lower electric fields, and the devices with doped transport layers. We show that these conditions, known to decrease the device degradation rates, may separately lead to the current homogenization in organic films.

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1. *Introduction*

Among organic electronic devices currently developed, the organic light-emitting devices (OLED) seem to be the first to have come to the market. The currently available organic flat displays appeared after a decade of struggle for increased device efficiency and durability. The initial discovery of small organic molecules [1] and polymers [2] suitable for efficient electroluminescence devices has been followed by numerous syntheses of improved materials. However, most of these materials still tend to degrade in prolonged contact with water and oxygen. Therefore, the first step in making more durable devices was to remove water and oxygen, as well as some other contamination sources, from the metal surface on which the organic materials were deposited, to control to the best degree their absence in the production process, and to protect the devices after production by using suitable encapsulating layers. Further steps were more involved and also required years of research. Partial improvement followed as new materials have been developed which are less susceptible to structural changes at elevated temperatures. On the other hand, rather soon after the first fairly efficient devices had been invented, the "Coulomb-ageing" rule was observed in OLED's [3]. This rule states that, for a given device structure, the degradation rate is proportional to the value of the initial luminosity L_0 (L_0 is proportional to the operating current), $\mathrm{d}L/\mathrm{d}t \approx -\gamma L_0 L$. A somewhat oversimplified interpretation of this observation suggests that each electron crossing the device produces an irreversible damage, thus limiting the overall light output during a device lifetime. This called for the constructing of more efficient devices (more photons per electron) and for the design of more stable materials. The more efficient devices were accomplished in a number of ways, from using the triplet emitters [4] to using multiple sandwich structures [5], leading to very durable devices. The increased efficiency allows for a lower operating current density and electric field for a desired light output. The devices operating at lower voltages (lower average electric fields) were also accomplished recently by using charge-doped organic layers [6]. However, the increase in the efficiency has not been the sole route to more durable devices. In a recent review, Popović and Aziz [7] summarize three additional modifications which separately lead to extended OLED lifetimes: the inclusion of a thin injection layer, doping of the transport layer by suitable organic molecules (trap doping), and mixing of the hole transport material and the electron transport material in the recombination region. Interestingly enough, and quite contrary to the mixed-heterojunction approach, the preparation of a smoother interface also results in more durable devices [8].

In this article, we consider theoretically most of the modifications enumerated above by studying their effect on the charge conduction through thin amorphous films. The study of the effect of material mixing on recombination is beyond the scope of the present paper. As for the modifications addressed here, we find that each of them may contribute to the homogenization of the current flow. While we do not consider the precise microscopic electronic mechanisms responsible for the device degradation, our findings suggest that the degradation may be closely related to inhomogeneous current flow in organic films.

We would like to point out here that Coulomb-ageing rule supports this suggestion. Scaled down from the full device to smaller "subdevices", the rule suggests the faster degradation rate for inhomogeneous devices. Indeed, the parts of a device that suffer a higher current density are expected to degrade faster. Since they also carry more current than the rest, their degradation affects the whole device more than the degradation of the other parts. The easiest way to demonstrate this mathematically is to consider a toy model of a device composed of two devices of equal surfaces but with unequal characteristics, connected in parallel and both degrading according to the same Coulomb-ageing rule. Given different current densities and different initial luminosities $L_{10} = L_0\alpha$ and $L_{20} = L_0(1-\alpha)$, the total luminosity then decreases according to

$$L(t) = L_1(t) + L_2(t) = L_0\alpha \exp\{-t\gamma L_0\alpha\} + L_0(1-\alpha)\exp\{-t\gamma L_0(1-\alpha)\}. \quad (1)$$

It is immediately seen that the diminution rate is the lowest for the symmetrical (homogenous) case, $\alpha = 1/2$, and increases as the asymmetry increases.

In reality, there are several reasons to expect the current distribution in disordered organic thin films to be very inhomogeneous. While the experimental support for this is still incomplete, several reasons for inhomogeneous current flow have fairly sound theoretical background. In the next section, we shortly review several possible sources of the inhomogeneity that have been recently discussed in the literature. Further sections are then devoted to numerical simulations of the hopping transport in disordered molecular media, particularly focused on the inhomogeneity of the current flow.

2. *Possible sources of current filamentation*

2.1. *Electrode roughness and imperfections*

The current injection from an electrode into a semiconductor is not perfectly homogenous in real systems when the electrode contains imperfections. While various kinds of imperfections are possible if the electrode is not chemically clean, the imperfections are still rather common even for chemically clean surfaces. The roughness of a metallic surface leads to the variation of the injection field and thus to the spatial variation of the injected current density. The degree of the current inhomogeneity depends on the electrode roughness as well as on the injection characteristics of the contact. In devices based on amorphous thin organic films, the strongly nonlinear injection characteristics and rather strong operating fields ($F \sim$ 1 MV/cm) imply that this inhomogeneity may be very high. A typical variation of the height of the electrode surface of the order of 10 nm over the distance of 100 nm may lead to a current density that is several hundred times bigger than the average [9]. Thus the electrodes that are not ideally flat introduce seeds for high-current-density filaments in the bulk of the organic material. These current inhomogeneities are reduced farther away from the electrode by the carrier diffusion in the direction perpendicular to the electric field. However, at operational fields of the order of 1 MV/cm, the spatial scale required for the homogenization is bigger than the usual organic film depths ($\sim$ 100 nm), leading to the filaments that cross the entire device. From the experimental side, the relation between electrode roughness and device degradation has been recently emphasized by several authors [10–12].

2.2. *Inhomogeneous Schottky barrier*

Recently, we pointed out that a strong filamentation of current is present even for ideally clean and flat electrodes [13]. We found that a strongly inhomogeneous charge-carrier injection is intrinsic for organic materials with correlated energetic disorder.

This spatial correlation of site energies is considered essential for an understanding of the strong field dependence of the electron mobility in disordered organic molecular materials. While several physical origins of correlation have been proposed, the very existence of the spatial correlation in disordered organic materials and its influence on charge transport is rather well established. One of the sources considered was the influence of the dipolar potential (the molecules are not perfectly symmetrical), leading to the site-energy correlations that decay as the inverse distance in space. The observed dependence of the mobility on the electric field was derived analytically for this particular type of correlated energetic disorder. This type of correlations is considered in the present paper (see Ref. [13] for more details). The inhomogeneous injection, as shown in Ref. [13], is related to the variation of the Schottky barrier over the injection surface. The barrier itself is positioned within the organic molecular material, but usually not much farther than 10 nm from the metal surface. Its spatial variation is the consequence of the energetic disorder in the material that causes a random formation of the injection canyons in the energy landscape. For the energetic-disorder strengths [14] of the order of 0.05 eV, characteristically found in transport layers in organic light-emitting devices, the variation of the injected current density, $\delta j/j_{\rm avg}$, is estimated theoretically to the order of several hundred [13]. Figure 1 shows an example of the current distribution in space, as calculated for the hopping injection and transport in the disordered organic media.

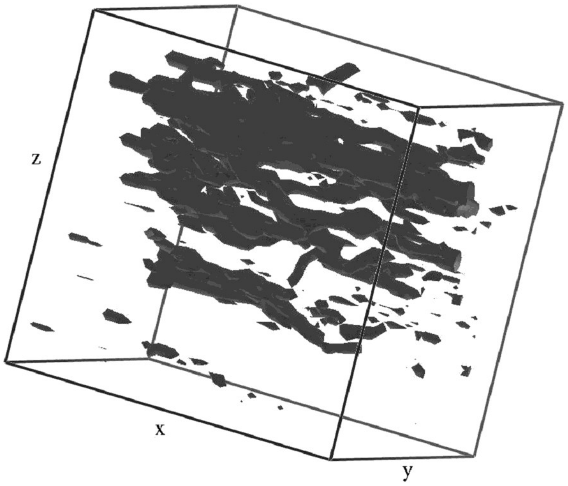

Fig. 1. The spatial profile of the current density in the system with correlated energetic disorder, $\sigma = 0.07$ eV, *homogenous in space [13]. The inhomogeneous injection is caused by the effect of disorder on the Schottky barrier, as described in Ref. [13]. The simulation of charge injection and transport shown here is done on the system with* $45\times45\times45$ *nodes (molecules). The electric field in the calculation is* $F = 0.5$ MV/cm. *The flat injection electrode is situated on the left side of the cube, while the exit electrode is on the right side.*

2.3. Rough organic heterojunctions

In practice, efficient light-emitting devices based on organic films almost regularly consist of several organic layers. Growing an organic heterojunction usually produces an interface with roughness ranging form several nanometers to tens of nanometers. Monopolar organic heterojunctions (i.e. those visited mostly by a single type of carriers, electrons or holes) may act as rough electrodes, provided the charge accumulation at the heterojunction is high enough to make the heterojunction an equipotential surface. However, even without calling for the Coulomb effects, the rough heterojunction barriers may seed current filaments as shown further on in this paper. It has been shown recently [8] that the evaporation of energetic molecular clusters is a good way to smoothen an organic heterojunction. In the line of the argument that we develop in this paper, it is interesting to note that this again leads to a multiple increase of the OLED lifetime [8].

3. The model and the method

The transport in disordered organic molecular media proceeds mostly by the phonon-assisted hopping of electrons among the electronic levels of neighboring molecules. These levels are energetically disordered even for a pure material, due to the Coulomb interaction among molecules and the structural disorder of the material. The distribution of energy levels E_i^0, usually taken as Gaussian in theoretical considerations [15], is characterized by its mean value E^{M}, related to the LUMO level of the molecules (or HOMO for hole transport), and its half-width σ. The latter depends on other parameters such as the degree of structural disorder and the strength of dipolar molecular moments. The correlation among E_i^0's in space, coming from the long-range nature of the dipolar interaction as well as from other sources, is regarded as the essential property of disorder in a pure material. The correlation is regarded responsible for the regularly observed Pool–Frenkel law for the charge mobility dependence on the electric field, $\log \mu \propto \sqrt{F}$ [16].

The hopping transport is described here by the master equation

$$\frac{\mathrm{d}n_i}{\mathrm{d}t} = \sum_{\delta} \nu_0 \left[n_{i+\delta} \mathrm{e}^{(E_{i+\delta}-E_i)/2T} - n_i \mathrm{e}^{-(E_{i+\delta}-E_i)/2T} \right], \tag{2}$$

where n_i stands for the probability of the i'th site being occupied, ν_0 represents the "bare" hopping rate among the sites, and $i+\delta$ runs over the nearest neighbors of the site. Both the injection and transport are described by this equation. The molecules next to the metal surface are taken to be in the equilibrium with the metal,

$$n_i = \mathrm{e}^{-(E_i^0-\mu)/T} \quad \text{for} \quad x_i = x_1, \tag{3}$$

while the injection is determined by the hopping rate over the Schottky barrier created, as usual, by the image-force potential and the external electric field, [17 – 19]

$$E_i = E_i^0 - \frac{q^2}{16\pi\varepsilon_0\varepsilon_r x_i} - Fq(x_i - x_1). \tag{4}$$

In this paper, we consider only the steady-state currents. We neglect the Coulomb interaction among carriers, taking its effect to be negligible as compared to the effect of the electric field produced by electrodes. Physically, this is acceptable when the charge density in the material is below the value that can produce sizable space-charge effects. This also means that we are basically confined to the low-current regime. Within these assumptions, the problem reduces mathematically to the linear set of equations

$$\sum_{\delta}\left[n_{i+\delta}\mathrm{e}^{(E_{i+\delta}-E_i)/2T}-n_i\mathrm{e}^{-(E_{i+\delta}-E_i)/2T}\right]=0\,, \tag{5}$$

where terms from Eq. (3) act as sources (inhomogeneous parts of the linear problem).

In spite of being a linear problem, solving for current paths in an inhomogeneous multidimensional system is, in general, still a rather demanding computational task. The technical difficulty is mainly related to the rank of the system of linear equations which is of the order of a million for a representative 3D problem, or of the order of ten thousand for a 2D problem with the comparable linear scale. However, one can obtain the solution due to the sparse matrix nature of the linear problem and recently developed efficient numerical methods for this type of mathematical problems [20].

The solutions for a number of cases are compared here; the transport in the pure disordered material is compared to those where different types of materials are combined. In the latter cases, two materials are characterized by different values of the mean value of E^{M} of molecular energy levels. For simplicity, the bare hopping frequency ν_0 is kept the same throughout the sample.

The problem of injection and transport in a pure disordered material has been recently addressed in Ref. [13]. The phenomenon discussed there is qualitatively the same in 3D and 2D systems: the essential physics of canyon forming in the injection area and the current dispersion is present in a random system as soon as one goes beyond 1D. Therefore, for the reason of computational efficiency, here we consider mostly the 2D systems. Also, much easier than 3D systems, the 2D systems lend themselves to graphical illustrations of the current flow, which we hold important for conveying our results. In order to check for the statistical significance of the results in our calculations, we consider several sets of independently generated energy configurations. For 2D simulations, each set consist of N two-dimensional systems, which are themselves of the size $N \times N$. Here we present the results for $N = 64$.

4. *The current homogenization by ordered injection layers*

Given the effect of disorder on the variation of the Schottky injection barrier, it is natural to expect that a semiconductor with less disorder may provide a more homogenous injection. The thickness of the ordered layer may be rather small, of the order of 10 nm, since this is the region where the Schottky barrier is placed. The related numerical simulations, confirming this, are illustrated in Fig. 2. The current is significantly homogenized inside the organic bulk. The remaining inhomogeneities can not be removed since they are not caused by the injection, but rather by the disordered nature of the bulk molecular material.

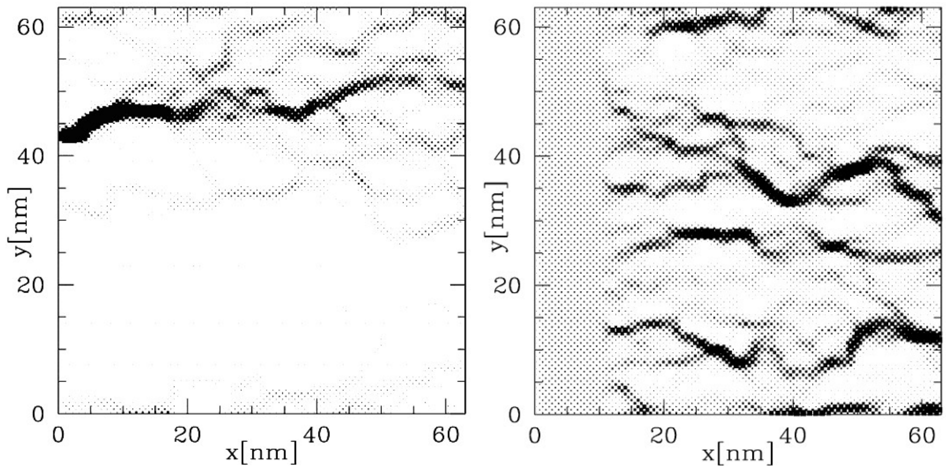

Fig. 2. The effect of an ordered injection layer on the current distribution at $F = 0.5$ MV/cm. *We compare two devices with the same disorder strength in the bulk,* $\sigma = 0.07$ eV. *In the second system a thin (10* nm*) injection layer without disorder is introduced. This layer is placed next to the injection electrode (on the left side of the sample). There is no energy barrier imposed at the organic heterojunction at* $x = 10$ nm *since the two materials are characterized by the same average value of the energy level. The current flows in both systems are normalized to the same total current, and the scales of gray are adjusted to the respective ranges of the current density. It may be seen that in the first system the current is concentrated in a single current filament. The introduction of the thin injection layer in the second system makes the current much more homogenous.*

It should be noted that the homogenization mechanism discussed here is different from the one previously discussed in Ref. [9]. There we considered the homogenization that is related to the smoothening of a metallic surface by an additional organic layer. A significant energy barrier and charge accumulation at the organic heterojunction was required there in order to produce a new "quasi-metallic" injection interface. No barrier like that, no significant carrier accumulation and no Coulomb interaction effects are assumed here. Reduced disorder in the vicinity of the metal surface was the sole means used here to homogenize the charge injection. It should be noted that reduced disorder relates in general to the higher carrier mobility in organic molecular material [16], $\ln \mu_0 \propto (\sigma/k_\mathrm{B}T)^n$, with $n \sim 2$. Indeed, the carrier mobility in copper-phtalocyanine (CuPc) used for thin injection layers is at least an order of magnitude higher that mobility in materials used for the bulk of the OLED devices [3]. It was also found that the optimal thickness of injection layers is of the order of 10 nm, just as required above to homogenize the Schottky injection barrier [21 – 23].

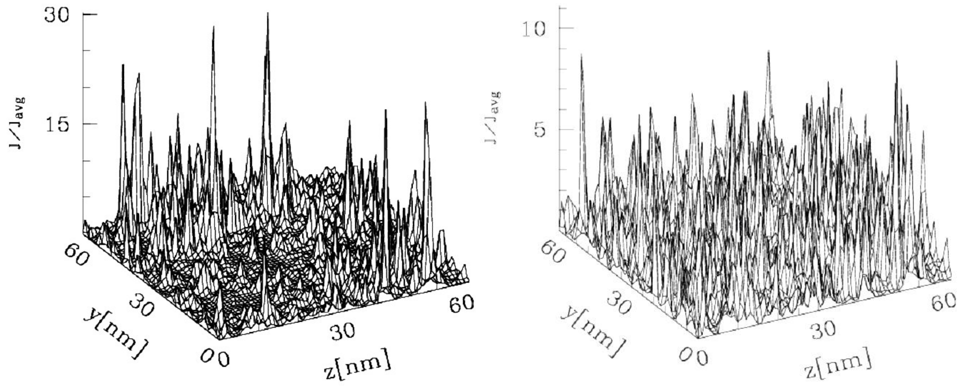

Fig. 3. This figure is related to the same data set as the previous one. It shows the current density distribution at the distance $x = 55$ nm *from the injection electrode, as obtained for the ensemble of 64 two-dimensional plates (i.e. 3D correlated energy landscape). The* $z = 35$ nm *cross-sections of these figures correspond to the cross-sections at* $x = 55$ nm *of Fig. 2. The homogenization of the charge flow caused by the ordered injection layer is visible by comparing the two figures.* $F = 0.5$ MV/cm.

5. *Operating the device at lower electrical fields*

As explained in the Introduction, the Coulomb-ageing rule calls for more efficient devices to attain improved durability. Indeed, some recent devices which went in that direction [4,5] operate at lower currents and lower voltages and reach the lifetimes far beyond 10000 hours, usually required for commercial applications. Although the reasons for high durability of those devices may go beyond reduced current filamentation, the latter is certainly characteristic of devices operating at lower voltages. To show this, we compare the current flows for low and high electric fields. The result is shown in Fig. 4. It may be observed that not only is the injection more homogenous for lower operational fields, but the current paths are less straight as well, leading to more dispersed current flow.

6. *The effect of organic heterojunction*

Next we consider the effect of a rough surface on charge transport. An example is shown in Fig. 5. For simplicity, the roughness is modeled by a sinusoidal curve with the amplitude of 4 nm. The separation between the neighboring peaks is taken to be 16 nm in order to have several peaks fitting within the sample width. A thin injection layer (Sec. 4) is first used to homogenize the current inflow. The organic heterojunction is then imposed farther inside the device, as indicated by the dashed

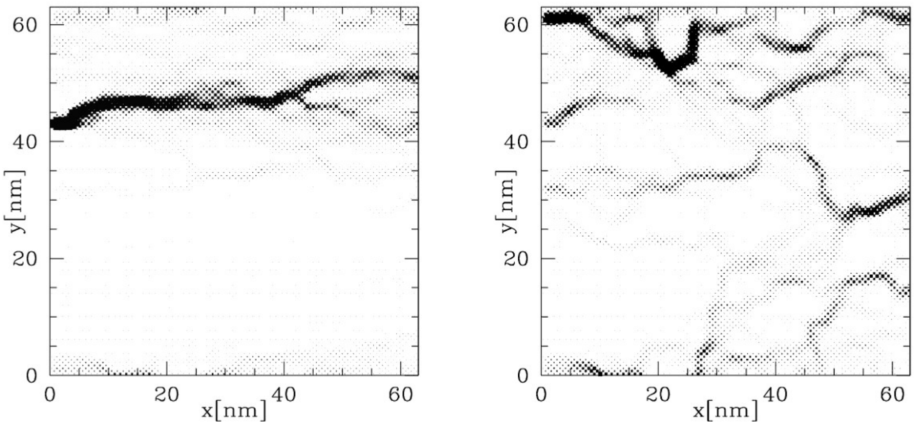

Fig. 4. The current density profiles for electric fields for $F = 1.0$ MV/cm *and* $F = 0.1$ MV/cm. *The current is much more inhomogenous for the higher field. At lower fields, the current density lines are much less straight and the meandering contributes to the homogenization of the current flow.*

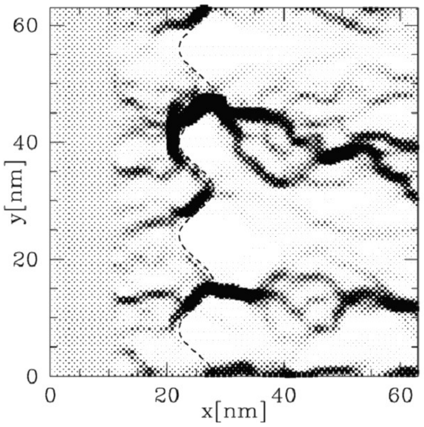

Fig. 5. The effect of a rough organic heterojunction barrier in the interior of the device. The dashed line indicates the heterojunction boundary. The two organic materials are parameterized by the same disorder strength, $\sigma = 0.07$ eV. *The energy barrier at the heterojunction is* 0.4 eV. $F = 0.5$ MV/cm.

line in Fig. 5. The figure shows that a rough molecular heterojunction may indeed represent an important seed for current filamentation in organic films.

The mechanism might seem similar to the one at a rough metal-organic interface. However, the difference is significant. The amount of charge accumulated at the interface and the mobility of electrons in the organic layer may be well below to make the interface an equipotential surface. The formation of the Schottky barrier, characteristic of the metal-organic heterojunction, is not likely for the same reasons. Indeed, no effects of the Coulomb interaction are taken into account in the calculation leading to Fig. 5. The filaments observed in the figure are produced by the effective sinks in the energy landscape. These sinks are created by the energy barrier, the surface roughness and the energetic disorder.

7. *The effect of doping on current homogenization*

Next we consider the effect of doping on current filaments. The results are not as unambiguous as those obtained in previous sections. It should be stressed that the doping we have in mind here is not the charge (donor/acceptor) doping, usual for electronic devices based on inorganic crystals. The effects of charge doping on the transport in organic disordered media, leading to lower fields needed to drive the same current, are probably qualitatively accounted for in Sec. 5. The doping mostly used in OLED's is the one that introduces traps or obstacles for the charge transport. The dopands are organic molecules whose HOMO and/or LUMO levels are above or below those of the organic host. These differences in energy levels are large enough to provide some trapping or barriers for electron transport, but not to induce spontaneous electronic transfer among different molecular species. In our model, the dopand sites are represented as electronic levels that are shifted with respect to the host matrix, $E_i^{0,\mathrm{dopand}} = E_i^{0,\mathrm{host}} + \delta E_d$, randomly positioned in space.

Figure 6 shows that preexisting current filaments may indeed be dispersed to some extent when they enter the doped region. However, we also find that deeply inside the doped region, and in particular without a strong preexisting filamentation, the intrinsic inhomogeneity in the current density may be higher than in an undoped system. This is indeed the consequence of the additional disorder in-

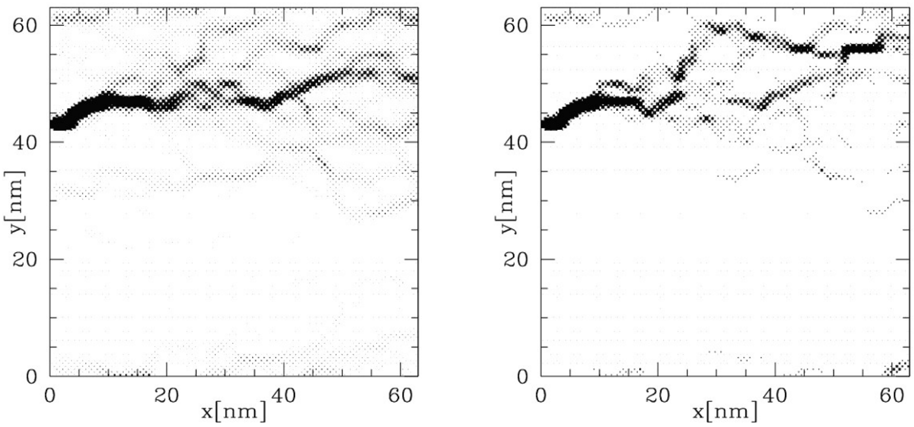

Fig. 6. The effect of doping with shallow-to-intermediate traps on the current dispersion. Although the effect is not very pronounced here, the doping (in panel b) introduces additional disorder in the bulk, which causes current filaments to scatter and disperse. Therefore, doping homogenizes the current distribution in a strongly filamented current flow. As in the previous figure, the two systems here are normalized to the same total current. The doping concentration is 8%. The doping is introduced for $x > 10$ nm *in the sample represented by the panel b). On the average, the traps are* -0.3 eV *below the average energy of the host.* $F = 0.5$ MV/cm.

troduced by doping. Increased disorder introduces a more inhomogeneous current flow on the microscopic level. We find similar effects of filament dispersing, followed by a more inhomogeneous flow on the finer spatial scale, both for doping by traps ($\delta E_d < 0$) and obstacles ($\delta E_d > 0$).

8. Conclusions

The numerical simulations in recent years have elucidated the nature of the strong field and temperature dependence of the mobility in disordered organic molecular media. The electronic conduction proceeds through thermally-assisted electron hopping within the manifold of electronic states that are disordered but correlated in space. We have used the same model to investigate another important aspect of the electronic conduction in disordered organic films – the filamentation of current. Several mechanisms are enumerated as probable causes of the filamentation. The previously formulated assumption that the filamentation may accelerate device degradation is given additional support in this paper. However, the experimental support for the scenarios discussed here is still missing. While probing current on the microscopic scale in the thin film buried between two electrodes is not an easy experimental task, there may be several ways to get valuable insights. For example, the experiment on the current distribution collected by the STM tip playing the role of one electrode above the organic-on-the-metal structure is desirable.

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USPOSTAVLJANJE STRUJNIH STAZA I POGORŠANJE SVOJSTAVA ELEKTRONSKIH SKLOPOVA ZASNOVANIH NA AMORFNIM ORGANSKIM SLOJEVIMA

Nedavni uzlet u proizvodnji organskih zaslona uslijedio je nakon duge borbe za produljenje vijeka trajanja tih uređaja. Mnoga su poboljšanja pri tom nastala kao rezultat eksperimentalnih pokušaja i promašaja. U ovom članku pokazujemo da su najvažnije nedavne preinake vrlo vjerojatno mikroskopski povezane s prostornim ujednačavanjem električne struje. Pri tom smo najprije ponovili glavne poznate razloge za pojavu prostornog uspostavljanja strujnih staza, pojave svojstvene za injekciju i vođenje u tankim organskim amorfnim slojevima. Potom smo numeričkim simulacijama redom razmotrili utjecaje: tankog injekcijskog sloja velike elektronske pokretljivosti; zaglađene naspram hrapave granice organskih slojeva; slabog nasuprot jakog operativnog električnog polja, te utjecaj punjenja na transport u organskim transportnim slojevima. Pokazujemo da sve te preinake u strukturi i načinu rada, znane da dovode do produljenja vijeka trajanja uređaja, također uzrokuju prostorno ujednačavanje struje u tankim slojevima.