Transport and excess noise in polyacenes under trap filling transition

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Abstract. Recent measurements in organic semiconductors (polyacenes) have evidenced a strong superlinear increase of the current-voltage characteristics associated with a sharp peak of the relative spectral density of current noise. Both features occur at the trap filling transition (TFT), i.e. at voltages corresponding to the transition between the Ohmic regime and the space charge limited current (SCLC) regime. Here we discuss the interpretation of this behavior in terms of a contribution from trapping and detrapping (TD) processes of the injected carriers. We found an excellent agreement between the predictions of our model and the experimental results in tetracene, which allows us to conclude that TD processes are responsible for the significant enhancement of the relative current noise observed in polyacenes in the TFT region.

1. Introduction
Organic devices, based on polymeric materials or molecular semiconductors, successfully compete with traditional electronic devices at least in terms of cost, flexibility and weight. The performance of organic devices is crucially controlled by the charge carriers injected at the molecule-metal interfaces. In turns, injected carriers are drastically affected by the presence of trapping centers related to defect states. The effect of thermal and electrical stresses on charge carrier trapping and detrapping processes was widely investigated in the literature [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. A smaller number of studies was devoted to noise in organic devices [12, 13].

In particular, recent transport and noise measurements in polyacenes [12] have evidenced a strong superlinear increase of the current-voltage, \( I-V \), characteristics associated with a sharp peak of the relative spectral density of current noise. This noise peak occurs at voltages corresponding to the crossover between Ohmic and space charge limited currents (SCLC) regimes [12], at the so called trap filling transition (TFT). The interpretation of these experiments was addressed in terms of a continuous percolation between a Ohmic phase and an insulating (SCLC) phase [12]. According to this model, the noise peak was attributed to the clustering of insulating regions among the conducting material, bringing a narrowing of the current paths inside the film and thus a strong increase of current noise.

Here we discuss an alternative interpretation of the observed behavior in terms of TD processes of the injected carriers by deep traps. The phenomenological model considered here makes use of the information on the ionized traps fraction, extracted from the experimental I-V characteristics. To validate the model we consider the measurements performed on tetracene.
thin films [12]. Moreover, we will also report the results of numerical simulations based on a new percolation model which further support these conclusions.

2. Model and Results

We start by considering the $I - V$ characteristic measured [12] at 300 K for a tetracene thin film as reported in Fig. 1 (log-log plot). The film, of thickness $L = 0.65 \mu m$ and cross-sectional area $A = 0.1 \text{cm}^2$, was sandwiched between Au and Al electrodes [12]. Charge transport is assumed to be mainly driven by holes injected from the Au contact. At increasing voltage, the characteristics show three regimes: Ohmic ($I - V$), TFT (strong superlinear $I - V$), SCLC (quadratic $I - V$). Now we assume that the current can be written as the sum of two asymptotic contributions, Ohmic and SCLC, respectively denoted as $I_{\Omega}$ and $I_{SCLC}$, where the last contribution is modulated by the fraction of filled (ionized) traps $u(V)$. Thus we take:

$$I(V) = I_{\Omega}(V) + u(V)I_{SCLC}(V) \quad (1)$$

where: $I_{\Omega}(V) = (n_0e\mu A/L)V$, $e$ is the elementary charge, $n_0$ and $\mu$ respectively are the average density and the mobility of the free thermal carriers, $I_{SCLC}(V) = (9Ae_0\varepsilon \varepsilon_r \mu \Theta /8L^3)V^2$, with $e_0$ and $\varepsilon_r = 3.5$ the vacuum permittivity and the relative dielectric constant of the material. The trapping parameter $\Theta \equiv n/n_t = (n_v/gn_t) \exp[-E_t/k_BT]$ is the ratio between the average densities of free, $n$, and trapped, $n_t$, carriers ($n_v$, $g$, $E_t$, $k_B$ and $T$ respectively are the valence band density of states, the trap degeneracy, the trap energy level, the Boltzmann constant and the temperature).

![Figure 1](image1.png)

**Figure 1.** Black (red in color view) squares: Current-voltage characteristic of a tetracene thin film measured at 300K [12]. The two lines show the slopes of the I-V at the lowest (dashed) and highest (dot-dashed) voltage regions. The solid curve fits the data with the expression given in the figure (see text for details).

![Figure 2](image2.png)

**Figure 2.** Fraction of filled traps vs voltage. The function $u(V)$ is obtained by inverting Eq.(1) and taking for $I$ the experimental values, while $I_{\Omega}$ and $I_{SCLC}$ are evaluated by considering respectively the low and high voltage limit. The two arrows point out the transition regions of the ohmic regime ($u \approx 0$.) and of the SCLC regime ($u \approx 1$.).

The function $u = u(V)$ is obtained by best fitting the experimental curve. The fit is shown in Fig. 1 while Fig. 2 reports the function $u(V)$. From the fit we find: $L/(Ae_0n_0) = 3 \times 10^{11} \Omega$ and $9Ae_0\varepsilon \varepsilon_r \mu \Theta /8L^3 = 6 \times 10^{-11} \text{A/V}^2$. Combining these results, we get for the effective mobility: $\mu_{eff} = \Theta \mu = 4.7 \times 10^{-10} \text{cm}^2/\text{Vs}$, and $n_0\mu = 1.4 \times 10^6 \text{(mVs)}^{-1}$. In absence of an independent determination of $\mu$ or $\Theta$, we can only discuss the possible range of values for the
relevant parameters of interest. In particular, by assuming \( \mu = 1 \text{ cm}^2/\text{Vs} \), a typical value for a single crystal, we find \( \Theta \approx 10^{-10} \) and \( n_0 \approx 10^4 \text{ cm}^{-3} \). By using the theoretical expression for \( \Theta \) reported above and by taking \( g = 2 \), \( n_v = 10^{21} \text{ cm}^{-3} \), \( n_t = 10^{13} \div 10^{18} \text{ cm}^{-3} \) we find \( E_1 = 0.7 \div 1.0 \text{ eV} \). We note that these values compare well with the values reported in the literature.

Figure 3 displays on a log-log plot the relative current noise spectral density, \( S(f, V) \equiv S(f, V)/I^2 \), measured at a frequency \( f = 20 \text{ Hz} \), in constant-voltage conditions, as a function of the applied voltage, for the same tetracene sample used to get the \( I - V \) in Fig. 1 [12]. Consistently with the interpretation of the \( I - V \) characteristics, we identify three regimes in the behavior of \( S(f, V) \): Ohmic, TFT and SCLC. Accordingly we write:

\[
S(f, V) = S_\Omega(f) + S_{TFT}(f, V) + S_{SCLC}(f, V)
\]

(2)

where: \( S_\Omega(f) = (1/ALn_0)(\alpha_\Omega/f) \), with \( \alpha_\Omega \) the Ohmic Hooge parameter [14] (as well known, the Ohmic component is independent of voltage). On the other hand, according to Ref. [14], the SCLC component can be written as: \( S_{SCLC}(f, V) = (4eL/5A\alpha_\Omega e \Theta)(\alpha_{SCLC}/f)(1/V) \), with \( \alpha_{SCLC} \) the SCLC Hooge parameter. Finally, for the TFT regime we use the standard expression for the relative trapping-detrapping noise with a lorentzian spectrum:

\[
S_{TFT}(f, V) = \frac{1}{N_t[1+(2\pi f \tau)^2]} u(V)[1-u(V)] = Bu(1-u)
\]

(3)

where \( N_t \) is total number of traps inside the volume of the device and \( \tau \) the carrier lifetime. The parameter \( B \) is chosen to reproduce the total excess noise measured at the given frequency in the TFT regime. This is obtained for \( B = S_{max}/0.25 = 2.8 \times 10^{-10} \) s, where \( S_{max} \) is the maximum experimental value of the noise. Furthermore, by defining as \( V_{th} \) the threshold voltage for the onset of the SCLC regime and by taking into account that \( u \approx 0 \) in the Ohmic regime, we can rewrite \( S(f, V) \) at a given frequency and for \( V < V_{th} \) as:

\[
S(f, V) = \left[1-u(V)\right] \left[S_\Omega + Bu(V)\right]
\]

(4)

The values obtained from the above expression are reported in Fig. 3 as a solid line with grey (orange in color view) diamonds. Moreover, Fig. 3 also shows as a solid grey (green) line the power-law fit with slope \(-1\) to the experimental data in the high voltage region. Thus, Fig. 3 points out an excellent agreement between the predictions of our model and the experimental results in tetracene, which allow us to state that TD processes are responsible for the significant enhancement of the relative current noise observed in polyacenes in the TFT region.

This statement is further supported by the results, reported in Fig. 4, of numerical simulations based on a new percolation model. The model considers a two-dimensional square-lattice, binary resistors network [15] (size \( N \times N \)), where each elementary resistor \( r_{i,j} \) is subjected to random transitions from two states of resistances \( r_1 \) and \( r_2 \), with probabilities:

\[
W_{1\rightarrow 2} = \exp[-(E_2 - qv_{i,j})/k_BT]
\]

and

\[
W_{2\rightarrow 1} = \exp[-E_1/k_BT],
\]

where \( v_{i,j} \) is the voltage applied to \( r_{i,j} \). \( E_1 \), \( E_2 \) are two activation energies and \( q \) is an effective charge. It should be noted that the relative current noise of the network depends only on its size \( N \) and on the three ratios: \( f_r = r_1/r_2 \), \( a = (E_2 - E_1)/k_BT \) and \( b = q/K_BT \). The results reported in Fig. 4 are obtained by assuming constant-voltage boundary conditions and by taking the following values for the parameters: \( N = 75 \), \( f_r = 290 \), \( a = 2.5 \) and \( b = 210 V^{-1} \).

In conclusion, we have developed a phenomenological model which provides a quantitative interpretation of the current-voltage characteristics and of the relative current noise in tetracene as function of the applied voltage. The excellent agreement with the experiments allow us to state that the sharp peak of noise in the TFT region basically arises from the fluctuating
occuancy of the traps due to TD processes. Furthermore, we have reported results of numerical simulations based on a new percolation model which furtherly support this conclusions. Finally, we remark that the measured current noise spectrum was found to be $1/f$-like rather than Lorentzian. However, the broadening of the trap level energy could account for the $1/f$-like power spectrum [14, 15].

2.1. Acknowledgments
The support of D. Kotowski, B. Kutrezba-Kotowska and M. Tizzoni at the early stage of the work is gratefully acknowledged.

3. References
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