Approaching the Trap-Free Limit in Organic Single Crystal Field-Effect Transistors

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Crystalline organic semiconductors, bonded by weak van der Waals forces, exhibit macroscopic properties that are very similar to those of inorganic semiconductors. While there are many open questions concerning the microscopic nature of charge transport, minimizing the density of trap states (trap DOS) is crucial to elucidate the intrinsic transport mechanism.

We explore the limits of state-of-the-art organic crystals by measuring single crystalline rubrene field-effect transistors that show textbook like transfer characteristics, indicating a very low trap DOS. Particularly, the high purity of the crystals and the very clean interface to the gate dielectric are reflected in an unprecedentedly low subthreshold swing of 65 mV/decade, remarkably close to the fundamental limit of 58.5 mV/decade.

From the measured subthreshold behavior we have consistently quantified the trap DOS by two different methods, yielding an exceedingly low trap density of $D_{\text{tr}} = 1 \times 10^{11} \text{cm}^{-2}\text{eV}^{-1}$ at an energy of $\sim 0.62 \text{eV}$. These numbers correspond to one trap per eV in $10^8$ rubrene molecules. The equivalent density of traps located at the interface is $D_{\text{it}} = 3 \times 10^{15} \text{cm}^{-2}\text{eV}^{-1}$ which puts them on par with the best crystalline SiO$_2$/Si field-effect transistors.

Keywords: Trap density of states, subthreshold swing, rubrene, Cytop

I. INTRODUCTION

Charge transport in semiconductors is strongly influenced by the presence of traps, energetically located in the band gap between the two transport levels. The quantification of the density of these trap states (trap DOS) is a crucial step towards understanding the electrical properties of the materials. There are various ways to determine the trap DOS experimentally, which range from photoelectron spectroscopy to direct measurements of the transport properties of a semiconductor device. A field-effect-transistor (FET) is a well suited device to study the trap DOS, as the spectral distribution of charge traps can be studied through changing the Fermi level by applying a bias to the gate contact. This method has become a powerful tool to study material properties in the field of organic semiconductors.

In the past 3 decades, organic field-effect-transistors (OFET) have come a long way from the first organic thin-film-transistor (OTFT) to single crystal OFETs with mobilities surpassing 40 cm$^2$/Vs. The first generation of OFETs did not have any clearly distinguishable subthreshold regime, where an exponential dependence of the drain current on the applied gate voltage is expected. Through advancements in thin-film deposition methods it has been possible to build OTFTs with a well pronounced subthreshold regime, comparable to inorganic amorphous transistors. High purity single crystals of organic molecules led to OFETs with a steeper subthreshold slope. To unleash the full performance of these single crystals, a compatible gate dielectric is required, which does not introduce additional charge traps in the semiconductor. More recently it has been shown that either an air-gap structure or a fluorinated polymer results in high performance OFETs with a high mobility and a steep turn-on, enabling fast switching speed and low power consumption. To further improve the turn-on characteristics, there has been a focus on thin and high-$\kappa$ dielectric layers to increase the gate capacitance, leading to a lower subthreshold swing.

In this study we focus on the subthreshold regime from a microscopic perspective, especially its relation to the trap DOS. Single crystal OFET measurements with extremely low subthreshold swing are presented and the theoretical description of the subthreshold regime to extract the trap DOS is summarized. Furthermore, a method is derived to estimate the Fermi energy at the turn-on voltage and thus the depth of these traps. The range of validity of this analysis is assessed using a full numerical simulation. With either method, we consistently found an extremely low density of deep trap states for rubrene, as low as in crystalline inorganic semiconductors.

II. RUBRENE SINGLE CRYSTAL FETS

We have built a series of rubrene single crystal FETs in a bottom-gate/bottom-contact configuration with the amorphous fluoropolymer Cytop (Asahi glass, Bellex Int. Corp.) as gate dielectric and a structured gate of evaporated Cr/Au on a Si/SiO$_2$ substrate. Evaporated Au was used as source and drain electrodes. The rubrene single crystals have been grown by physical vapor transport from 98% pure source material (Sigma-Aldrich) without any additional purifying steps and were attached to the prefabricated substrates by flip crystal technique.

All devices show very similar electrical characteristics: Mobilities extracted from the saturation and the linear regime range from 10 to 15 cm$^2$/Vs and on-off ratios...
above $10^7$ at $V_g = -10$ V are reached. A common feature is the extremely steep turn-on behavior with a subthreshold swing $S$ in the range of 65 to 80 mV/decade. None of the devices show any hysteresis, i.e. no gate bias stress, implying that there is no long-term charge trapping in the OFET. The gate leakage current is below the noise level of the measurement setup at 200 fA. In the linear regime, the drain currents scale linearly with the gate voltage, which is a sign of a negligible small charge injection barrier at the contacts.

Here, we discuss the transistor with the lowest subthreshold swing (Fig. 1). The rubrene crystal was laminated in ambient air and measured in helium atmosphere at room temperature ($T = 295$ K), using a HP 4155A semiconductor parameter analyzer operated with instrument control (iC).

The transfer-curves for various drain voltages and the output characteristics are shown in Fig. 2 and 3. The 2-point field-effect mobilities derived from the linear and the saturation region, $\mu_{\text{lin}} = 13.0$ cm$^2$/Vs and $\mu_{\text{sat}} = 13.9$ cm$^2$/Vs, are remarkably high. In the following we focus on the subthreshold regime which is defined as the region between the turn-on and the threshold voltage, in this device given as $V_{\text{on}} = 0.47$ V and $V_{\text{th}} = 0.23$ V (from saturation regime). In this regime, the drain current increases exponentially with the gate voltage, which is defined as the subthreshold slope (in units decade/V) or its inverse, the subthreshold swing $S$ (V/decade). The latter is best extracted from a plot of the inverse logarithmic slope of the drain currents versus gate voltage (Fig. 4). In this plot the extremely steep exponential turn-on behavior becomes apparent: In the subthreshold region (Fig. 4 inset), the curves truncate at a minimal value of $S = 65 \pm 2$ mV/decade, which is the subthreshold swing. The same value for $S$ was obtained from both sweep directions (no hysteresis) and lies remarkably close to the theoretical limit of $S = 58.5$ mV/decade at 295 K.

### III. SUBTHRESHOLD SWING AND TRAP DENSITY

In the subthreshold region, where the gate voltage is below the threshold voltage, the formation of a pinch-off zone with a very low charge carrier density near the drain contact leads to a suppression of the drift currents.
Fig. 4. Inverse logarithmic slope of the measured transfer curves from Fig. 2. In the main panel, linear and saturation regimes manifest themselves in the known way: $I_d \propto V_g$ and $I_d \propto V_g^2$, respectively. A magnification of the subthreshold region is shown in the inset. With increasing gate voltage, the inverse slope is approaching a subthreshold swing value of 65 mV/decade, very close to the theoretical limit of 58.5 mV/decade.

The large gradient of the charge concentration between the source and drain contact regions, however, gives rise to a diffusion current which is independent of the drain voltage, as long as the drain voltage is larger than a few $k_B T/q$. The subthreshold-current is proportional to the carrier concentration which varies exponentially with the gate voltage. Thus,

$$I_d \propto \exp \left( \frac{qV_g}{n^* k_B T} \right),$$

where the so-called subthreshold slope depends on the thermal energy $k_B T/q$ and the ideality parameter $n^*$. The subthreshold swing $S$ is defined as the inverse of the subthreshold slope and corresponds to the gate voltage needed to increase the drain current by a factor of 10,

$$S = \frac{k_B T \ln(10)}{q} n^* .$$

The ideality parameter $n^*$ is associated with the density of charge traps far away from the transport level, either located at the semiconductor-insulator interface or in the bulk of the semiconductor. Due to their relatively large trapping energy these states are called deep traps. The parameter $n^*$ can be written as

$$n^* = 1 + C_{sc}/C_i ,$$

where $C_i$ is the capacitance of the gate dielectric per unit area. The quality of the semiconducting material expresses itself in an effective capacitance $C_{sc}$, since the filling of trap states while the Fermi energy is pushed towards the transport level is equivalent to the charging of a capacitor $C_{sc}$ is distinct from the geometric capacitance of the semiconductor, $C_{geom} = \varepsilon_{sc}/t_{sc}$. While $C_{geom}$ does not express itself in the DC transfer characteristics, $C_{sc}$ affects the subthreshold swing. Rolland et al. have shown it to be directly dependent on the density of deep trap states in the bulk and at the interface,

$$C_{sc} = q \sqrt{\varepsilon_{sc} D_{bulk} + q^2 D_{it}} ,$$

where $D_{bulk}$ is the bulk trap density per volume and energy, $D_{it}$ denotes the interface trap density per unit area and energy and $\varepsilon_{sc}$ is the permittivity of the semiconductor material.

For an ideal transistor without any traps, $C_{sc}$ is zero and the parameter $n^*$ equals 1. Thus, there is a theoretical limit for the subthreshold swing, given by $S_{ideal} = k_B T \ln(10)/q$ which is 58.5 mV/decade at 295 K.

For a real semiconductor, however, the subthreshold swing is larger than this minimum and the difference between the theoretical minimum and the measured subthreshold swing is a measure of the imperfection of the transistor interface and the semiconductor material.

From the subthreshold swing alone it is not a priori possible to distinguish between trapping at the interface and trapping in the depletion zone of the bulk, since both contribute to an effective capacitance $C_{sc}$, corresponding to a trap concentration per unit area. However, we can estimate the maximum density of interface traps contributing to the measured subthreshold swing by setting $D_{bulk}$ to zero:

$$D_{it}^{\text{max}} = \frac{C_{i}}{q^2} \left( \frac{q S}{k_B T \ln(10)} - 1 \right) .$$

If at least part of the trap states are located in the bulk, a reasonable assumption for the channel thickness is necessary to convert the areal density into a volume density. In general, this thickness is given by the Debye length $\lambda = \sqrt{\varepsilon_{sc}/q^2 D_{bulk}}$, which was also used in the derivation of eq. (5). Again, by setting $D_{it} = 0$ we obtain the maximum contribution of bulk traps:

$$D_{it}^{\text{max}} = \frac{C_{i}^2}{\varepsilon_{sc} q^2} \left( \frac{q S}{k_B T \ln(10)} - 1 \right)^2 .$$

Care must be taken when the above relations are applied to nearly trap-free semiconductors or to very thin semiconducting layers as in evaporated or solution-processed FETs. For such devices $\lambda$ can be of the same order as the semiconductor thickness $t_{sc}$ and the charge carriers accumulate almost uniformly throughout the semiconductor. In this situation, the charge transport in the subthreshold region is essentially a volume phenomenon rather than an accumulation of charge carriers within the first few monolayers of the semiconductor.

If the characteristic thickness of the conducting channel $\lambda$ is larger than $t_{sc}$, the relation (5) is no longer valid to extract the maximum bulk trap density. From the
measured subthreshold swing and the device geometry, one can directly assess if \( \lambda > t_{sc} \) by rewriting the Debye length \( \lambda \) in terms of \( S \):
\[
\frac{S}{S_{\text{ideal}}} < \frac{\varepsilon_{sc} t_{\text{ins}}}{t_{sc} \varepsilon_{\text{ins}}} + 1, \tag{7}
\]
where \( t_{\text{ins}} \) is the thickness and \( \varepsilon_{\text{ins}} \) the permittivity of the gate dielectric. Noteworthy, the borderline \( \lambda = t_{sc} \) is equivalent to the situation where the trap related effective capacitance \( C_{sc} \) equals the geometric capacitance \( C_{\text{geom}} \).

If the Debye length \( \lambda \) exceeds the crystal thickness \( t_{sc} \), the analysis of \( S \) in terms of trap density is slightly modified:
\[
C_{sc} = q^2 (D_{it} + t_{sc}D_{bulk}) \tag{8}
\]
and accordingly the maximum bulk trap density is
\[
D_{\text{bulk}}^{\text{max}} = \frac{C_{i}}{q^2 t_{sc}} \left( \frac{qS}{k_B T \ln(10)} - 1 \right). \tag{9}
\]

After this discussion it is clear that from a physical point of view the as measured subthreshold swing \( S \) is not suitable for a direct comparison between different FETs. Sometimes the subthreshold swing multiplied by the gate capacitance has been used for this purpose, but this value still depends on the geometry of the gate dielectric and gives no information about the intrinsic properties of the material. The key quantity to evaluate and compare the quality of the semiconductor and its interface to the di-electric is the capacitance of the transport channel \( C_{sc} \) obtained from \( S \) according to Eqs. (8) and (9).

**IV. ESTIMATION OF THE TRAPPING ENERGY**

Not only can we extract the density of trap states dominating the subthreshold region, but also their energy relative to the transport level using the following considerations:

The subthreshold current is dominated by diffusion and given by
\[
I_d = q W \lambda D \frac{P_{m,s} - P_{m,d}}{L}, \tag{10}
\]
where \( W \) and \( L \) are the width and length of the channel, \( \lambda \) is the channel thickness and \( D \) is the diffusion coefficient which is connected to the mobility \( \mu \) by the Einstein relation \( D = \mu k_B T / q \). Since in the subthreshold regime the concentrations of mobile charges near the source, \( p_{m,s} \), and the drain, \( p_{m,d} \), differ by several orders of magnitude, we set \( p_{m,d} = 0 \). Furthermore, if the Fermi level lies several \( k_B T \) away from the transport level, \( p_{m,s} \) is given by Maxwell-Boltzmann statistics:
\[
p_{m,s} = N_{\text{band}} e^{-\frac{E_{F} - E_{sc}}{k_B T}}, \tag{11}
\]
where \( N_{\text{band}} \) is the number of states in the transport level and \( \Delta E = E_F - E_{sc} \) is the Fermi energy relative to the transport level \( E_{sc} \).

Again, we consider the two situations where the characteristic channel thickness given by the Debye length \( \lambda = \sqrt{\varepsilon_{sc}/q^2 D_{\text{bulk}}} \) is either smaller or larger than the geometric thickness \( t_{sc} \) of the semiconductor. For \( \lambda < t_{sc} \), combining Eqs. (10) and (11) results in
\[
\Delta E = -k_B T \ln \left( \frac{q I_d L}{N_{\text{band}} W \mu k_B T} \sqrt{\frac{D_{\text{bulk}}^{\text{max}}}{\varepsilon_{sc}}} \right). \tag{12}
\]

and the density of bulk traps per unit energy \( D_{\text{bulk}}^{\text{max}} \) can be obtained from the subthreshold swing by using Eqn. (5). \( I_d \) is the drain current for which the minimal subthreshold swing is reached. If on the other hand the Debye length exceeds the thickness of the semiconducting layer \( \lambda > t_{sc} \) we obtain
\[
\Delta E = -k_B T \ln \left( \frac{I_d L}{N_{\text{band}} W t_{sc} \mu k_B T} \right). \tag{13}
\]

With equation (12) or (13) the Fermi energy and thus the energy of the deep trap states filled upon turn-on can be estimated directly from macroscopic values.

**V. NUMERICAL SIMULATION OF THE SUBTHRESHOLD CURRENT**

The analytical method discussed in the previous sections is convenient to estimate the density of deep trap states. Its derivation, however, is fairly involved and it is desirable to assess the validity of this model in detail. We performed a series of numerical simulations of the FET’s subthreshold current assuming a wide range of bulk trap densities and we have compared the resulting subthreshold swing with the values predicted by the analytical model (Eq. 5).

For the numerical calculations, a new in-house implementation of a FET model was used: It solves the drift- and diffusion equations in the presence of traps, similar to previous simulators. The differential equations are solved in 2 dimensions for the entire operating regime of the transistor and the trap DOS can be chosen arbitrarily. Methods to numerically solve these equations are discussed in more detail e.g. in the book *Analysis and Simulation of Semiconductor Devices*.

The drift- and diffusion equations for hole only transport consist of Poisson’s equation, the continuity equation and the definition of the drift and diffusion current density \( \vec{J}_p \):
\[
\vec{\nabla}^2 \Psi = -\frac{q}{\varepsilon_{sc}} (p_m + p_t) \tag{14}
\]
\[
\vec{\nabla} \cdot \vec{J}_p + q R = -\frac{\partial p_m}{\partial t} \tag{15}
\]
\[ \vec{J}_p = -q \left( \mu p_m \vec{\nabla} \Psi + D \vec{\nabla} p_m \right), \]  
where \( \Psi \) is the electric potential, \( p_m \) and \( p_t \) are the mobile and trapped charge carrier densities, respectively, \( R \) is the recombination rate (here \( R = 0 \)) and \( \mu \) denotes the drift mobility for holes. The diffusion constant \( D \) is directly connected to the mobility by the Einstein relation.

This system of non-linear differential equations is discretized into finite differences and solved for the steady-state (i.e. \( \partial p_m/\partial t = 0 \)) in two dimensions, using the Gauss-Newton algorithm.

Dirichlet boundary conditions are used to fix the potential and the charge carrier density at the injecting contacts. The interface to the insulating gate dielectric is determined by the Neumann boundary condition \( \vec{J}_p \parallel \vec{n} = 0 \), and the electric fields in the semiconductor and the insulator are connected by Gauss’ law,
\[ \varepsilon_{sc} \frac{\partial \Psi}{\partial n}_{isc} - \varepsilon_{ins} \frac{\partial \Psi}{\partial n}_{ins} = Q_d, \]  
where \( \vec{n} \) is the unit vector orthogonal to the interface plane and \( Q_d \) is the sheet density of additional charge at the interface. For the simulations discussed here we set \( Q_d = 0 \).

The concentration of mobile and trapped charge carriers is determined by a convolution of the density of states with the Fermi-Dirac distribution for holes,
\[ p_m(E_F, T) = \int D_{\text{band}}(E) \cdot (1 - f(E, E_F, T)) \, dE \]  
\[ p_t(E_F, T) = \int D_{\text{trap}}(E) \cdot (1 - f(E, E_F, T)) \, dE, \]  
where \( D_{\text{band}} \) and \( D_{\text{trap}} \) are the spectral distribution of band-like (mobile) states and traps, respectively. From Eqs. (16) and (17), a relation \( p_m(p_t, T) \) can be calculated for any arbitrary distribution of conducting states and traps.

For this study, the DOS model illustrated in Fig. 5(a) was used: A 0.3 eV wide constant band containing \( 3 \times 10^{11} \) states per cm\(^{-3} \) represents the mobile states in the highest occupied molecular orbit level (HOMO). Since we are focussing on the subthreshold region only, we assume for simplicity a constant trap DOS in the energy range relevant at the turn-on voltage. This is a reasonable approximation far away from the transport level, as has been measured in previous studies on single crystal and thin film OFETs, where the trap DOS changes by a factor of \( \sim 2 \) within a few \( k_B T \). We varied the trap density over several orders of magnitude and calculated the transfer characteristics in the subthreshold regime at different temperatures (example in Fig. 5(b)). From these curves we take the minimum subthreshold swing \( S \) and compare the values in Fig. 6 to the predictions by the analytical model (Eq. 9 and 10) respectively for the two cases, \( \lambda \lesssim t_{sc} \). First of all, we note the excellent agreement in the entire parameter space. Furthermore, the simulations confirm the need to distinguish between the two ranges (dashed line).
VI. RESULTS AND DISCUSSION

The above described analysis method is a powerful tool to probe the microscopic physical parameters of the rubrene single crystal FET (Fig. 2 and 3). These measurements are of particular interest because the calculated Debye length of $\sim 4 \mu m$ places this device in the parameter space where the channel thickness at turn-on is limited by the crystal thickness. The extracted results are summarized in Table I.

| param. | value | unit |
|--------|-------|------|
| $T$    | 295   | K    |
| $S$    | 65 $\pm$ 2 | mV/dec |
| $n*$   | 1.11 $\pm$ 0.03 | |
| $C_i$  | 4.71 $\pm$ 0.09 | nF/cm$^2$ |
| $C_{sc}$ | 0.52 $\pm$ 0.16 | nF/cm$^2$ |
| $\varepsilon_{sc,rel}$ | 3.5 | |
| $D_{\text{bulk}}^i$ | $(1.3 \pm 0.4) \times 10^{13}$ | cm$^{-3}$eV$^{-1}$ |
| $D_{\text{it}}^i$ | $(3 \pm 1) \times 10^9$ | cm$^{-2}$eV$^{-1}$ |
| $\Delta E$ | 0.62 | eV |

TABLE I. Subthreshold parameters extracted from the transfer characteristics of the measured rubrene single crystal FET.

Assuming all the measured trap states to be located at the interface, we calculate a $D_{\text{it}}^i$ of $3 \times 10^9$ cm$^{-2}$eV$^{-1}$. We compare this number to the number of rubrene molecules at the interface. The lattice parameters of an orthorhombic rubrene crystal are $a = 26.9 \, \AA$, $b = 7.2 \, \AA$, $c = 14.4 \, \AA$, leading to a density of $9.6 \times 10^{13}$ unit cells per cm$^2$ in the b-c-plane (according to the crystallographic axis-definition) and thus to a molecule density of $3.9 \times 10^{14}$ cm$^{-2}$. Therefore, the deep trap density per eV is less than $1$ in $10^5$ molecules. Alternatively, if we assume all the traps to be distributed throughout the bulk, the same reasoning leads to a density of defects electronically active far away from the transport level of $1$ in $10^8$ molecules per eV.

Considering the chemical purity of the source material (98%) the density of measured defects is remarkably low. This exemplifies the efficient purification during growth of the crystal by physical vapor transport. Furthermore, the equivalent interface trap density of only $D_{\text{it}} = 3 \times 10^9$ cm$^{-2}$eV$^{-1}$ compares very favorably with the best crystalline SiO$_2$/Si interfaces, where $D_{\text{it}}$ is in the range of $10^{10}$ cm$^{-2}$eV$^{-1}$.

The comparison of the analytical DOS extraction method with the numerical simulations reveals good general agreement in a broad range of bulk trap densities and temperatures (Fig. 3). Not surprisingly, the small deviations are most pronounced near the borderline at which we distinguish between the situations of a characteristic channel length shorter or larger than the actual semiconductor thickness. However, even in this region the difference in $D_{\text{bulk}}$ is at most half an order of magnitude, indicating that this method is well suited for OFETs with a nearly trap-free interface and bulk, or with a very thin semiconducting layer.

The calculated Fermi level is $\Delta E = 0.62$ eV (Eq. 13), at the voltage where the subthreshold swing and thus the trap density has been determined ($I_d = 3 \times 10^{-12}$ A). More generally, the deep trap energy probed in the subthreshold region depends on the device geometry and the lowest measured $I_d$ (limited by noise level or off-current). Therefore, optimizing these parameters gives access to even deeper trap states.

VII. CONCLUSION

The rubrene FETs in this study have an extremely low density of deep trap states, indicating the high quality of the semiconductor. Here, this is seen in an unprecedentedly low subthreshold swing of $S = 65$ mV/decade at room temperature, which lies remarkably close to the theoretical trap free limit at $58.5$ mV/decade.

An analytical method is shown to be well suited to experimentally access the density of deep bulk and interface traps from the subthreshold region of FET transfer curves. This method was verified by a comparison to numerical simulations of the subthreshold current. A novel way to estimate the trapping energies was presented, which additionally gives an upper limit of the subthreshold dominating energy range.

With this in-depth analysis we estimate a trap density of $D_{\text{bulk}} = 1 \times 10^{13}$ cm$^{-3}$eV$^{-1}$, or equivalently $D_{\text{it}} = 3 \times 10^9$ cm$^{-2}$eV$^{-1}$, at 0.62 eV above the transport level. This demonstrates that deep trap densities in the best organic single crystal FETs are as low as in the most advanced crystalline SiO$_2$/Si transistor. This may come as a surprise considering the straightforward fabrication method of the OFETs, including manual flip-crystal lamination in ambient air.

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