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Supplementary Materials for

Flexible low-voltage high-frequency organic thin-film transistors

James W. Borchert*, Ute Zschieschang, Florian Letzkus, Michele Giorgio, R. Thomas Weitz, Mario Caironi, Joachim N. Burghartz, Sabine Ludwigs, Hagen Klauk*

*Corresponding author. Email: j.borchert@fkf.mpg.de (J.W.B.); h.klauk@fkf.mpg.de (H.K.)

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S1: Contact resistance, carrier mobility and transit frequency

The contact resistance (R_c) in organic TFTs is typically evaluated in the linear regime of transistor operation using the transmission line method (TLM; see Figs. 2 and S4) (41). In TLM analyses, as well as for most other methods for determining $R_C(42)$, a requirement (and indeed a drawback) is that it is only valid for the linear regime of operation using vanishingly-small V_{DS} , since a key assumption of TLM is that R_C is Ohmic and that the channel resistance is uniform over the entire channel region. This latter point by itself precludes any analysis of R_C in the saturation regime using TLM because of the pinch-off of the channel that occurs when V_{DS} is approximately equivalent to the overdrive voltage (V_{GS} - V_{th}). Experimental investigations have shown that R_C in organic TFTs can vary significantly with V_{DS} (32, 43). Especially for TFTs with small channel lengths (L), R_C may be reduced by V_{DS} due to effects such as image-force lowering (IFL) (33) or by drain-induced barrier lowering (DIBL) (35) of the injection barrier at the interface between the source contact and the semiconductor. Enhancements of the carrier mobility in the vicinity of the contacts through a Poole-Frenkel-like dependence of the mobility on the applied electric field may also lead to lower R_{C} , in part due to the nonlinear effects on both charge transport (44) and charge injection (45). For these reasons it is beneficial to be able to quantify the dependence of R_C on V_{DS} .

Here, we estimate R_C and the intrinsic channel mobility (μ_0) in the saturation regime ($V_{DS} \leq V_{GS} - V_{th}$ for p-channel TFTs) by fitting the transit frequency (f_T) extracted from S-parameter measurements as a function of channel length (L). Similar to TLM, we use μ_0 and the width-normalized contact resistance (R_CW) as fitting parameters. This approach has to our knowledge never been explicitly implemented, though comparisons of calculated f_T to experimental results using R_C determined from TLM have been reported (16, 36). In both approaches, the error in the extracted values is assessed simply as the calculated standard error from the fits. In the TLM, the width-normalized total source-to-drain resistance of the TFT (RW) is fit with a linear function with respect to L. In our method, the f_T data as a function of L is fit using the equation derived in the following.

The dependence of the effective mobility (μ_{eff}) and the transit frequency (f_T) on R_C can be illustrated with the following two equations for TFTs operated in the saturation regime (2, 46):

$$\mu_{eff} = \mu_0 \left[1 - \left(\frac{\mu_0 C_{diel} R_C W(V_{GS} - V_{th})}{L + \mu_0 C_{diel} R_C W(V_{GS} - V_{th})} \right)^2 \right]$$
(1)

$$f_T = \frac{\mu_{eff}(V_{GS} - V_{th})}{2\pi L \left(L_{ov,total} + \frac{2}{3}L \right)}$$
(2)

where C_{diel} is the gate-dielectric capacitance per unit area and $L_{ov,total}$ is the total gate-to-contact overlap length, which is simply the sum of the gate-to-source ($L_{ov,GS}$) and gate-to-drain ($L_{ov,GD}$) overlap lengths. In principle, the term R_C encompasses contributions from both the source and drain contacts. Equation 1 can be simplified to (36)

$$\mu_{eff} = \frac{\mu_0}{1 + \frac{1}{2L} \mu_0 C_{diel} R_C W(V_{GS} - V_{th})}$$
(3)

Combining Equations 2 and 3, we then obtain an expression for f_T that includes the influence of the contact resistance:

$$f_T = \frac{\mu_0 (V_{GS} - V_{th})}{2\pi \left(L + \frac{1}{2} \mu_0 C_{diel} R_C W (V_{GS} - V_{th}) \right) \left(L_{ov,total} + \frac{2}{3} L \right)}$$
(4)

For illustrative purposes, several curves of the transit frequency as a function of the channel length calculated using Equation 4 and assuming different values for R_CW are shown in Fig. S7.

Supplementary Figures



Fig. S1| Device fabrication process and materials characterization. (A) Schematic process flow for the fabrication of bottom-gate bottom-contact (inverted coplanar) organic TFTs. All metal and semiconductor layers are deposited by thermal evaporation or sublimation in vacuum and patterned using high-resolution silicon stencil masks. (B) Infrared reflection absorption spectroscopy (IRRAS) analysis of bulk pentafluorobenzenethiol (PFBT, black) and of a chemisorbed monolayer of PFBT on a gold surface (red). (C) AFM height scan of a thin film of the organic semiconductor DPh-DNTT deposited onto a hybrid AlO_x/SAM gate dielectric on a flexible PEN substrate.



Fig. S2| **Static transistor characteristics and uniformity.** (**A**) SEM micrograph of an individual DPh-DNTT TFT. (**B**) Measured transfer characteristics of 10 nominally identical TFTs having a channel length (*L*) of 1.5 μ m, a total gate-to-contact overlap ($L_{ov,total}$) of 60 μ m and a channel width (*W*) of 7.5 μ m, with statistics for the effective carrier mobility (μ_{eff}), threshold voltage (V_{th}) and subthreshold swing (*SS*). (**C**) Transfer characteristics of an individual TFT measured at a drain-source voltage (V_{DS}) of -3 V. The dotted blue line is a guide to the eye, indicating the ideal quadratic dependence of the drain current on the gate-overdrive voltage (V_{GS}) from 0 to -3 V in steps of 0.5 V.



Fig. S3| Static characteristics of TFTs based on DPh-DNTT (top row) and C₁₀-DNTT (bottom row). Both TFTs have a channel length (*L*) of 8 µm, a total gate-to-contact overlap ($L_{ov,total}$) of 4 µm and a channel width (*W*) of 200 µm. From left to right: Transfer characteristics, effective carrier mobility (μ_{eff}) plotted as a function of the gate-source voltage, and output characteristics for gate-source voltages (V_{GS}) from 0 to -3 V in steps of 0.5 V. (Note that these TFTs were fabricated separately from the TFTs shown in Fig. S2.)



Fig. S4| Transistors for TLM analysis. (A) Transfer characteristics of DPh-DNTT TFTs with channel lengths (*L*) ranging from 1 to 10.5 µm and a channel width (*W*) of 50 µm, measured with a drain-source voltage (V_{DS}) of -0.1 V. (B) Effective carrier mobility (μ_{eff}) in the linear regime determined for each TFT as a function of the channel length (*L*). The line is a fit to the data using the equation $\mu_{eff} = \mu_0 (1 + L_{1/2}/L)^{-1}$ where μ_0 is the intrinsic channel mobility and $L_{1/2}$ is the channel length at which $\mu_{eff} = \frac{1}{2} \mu_0$. (C) SEM micrographs of the channel region of the TFTs. (Note that these TFTs were fabricated separately from the TFTs shown in Fig. S2.)







	— <i>L</i> = 1.1 μm	
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Fig. S5| **Transistors for** *S*-**parameter measurements.** (**A**) Transfer characteristics of DPh-DNTT TFTs with channel lengths (*L*) ranging from 0.7 to 10.5 μ m, a total gate-to-contact overlap ($L_{ov,total}$) of 10 μ m and a channel width (*W*) of 100 μ m, measured with a drain-source voltage (V_{DS}) of -3 V. (**B**) Output characteristics of each TFT for a gate-source voltage (V_{GS}) of -3 V. (**C**) Channel-width-normalized peak transconductance (g_m) at a gate-source voltage (V_{GS}) of -3 V as a function of the inverse of the channel length. (**D**) SEM micrographs of the channel region of the TFTs. (Note that these TFTs were fabricated separately from the TFTs shown in Fig. S2.)



Fig. S6| DPh-DNTT TFT showing a transit frequency of 21 MHz. The TFT has a channel length (*L*) of 0.6 μ m, a total gate-to-contact overlap ($L_{ov,total}$) of 10 μ m and a channel width (*W*) of 100 μ m. (A) Output characteristics for gate-source voltages (V_{GS}) from 0 to -3 V in steps of 0.5 V. (B) SEM micrographs of the channel region of the TFT.



Fig. S7| **Relation between channel length, contact resistance and transit frequency.** The curves were calculated using Equation (4) for width-normalized contact resistances (R_CW) of 10, 50 and 100 Ωcm, gate-to-source and gate-to-drain overlaps ($L_{ov,GS}$, $L_{ov,GD}$) of 5 µm, an intrinsic channel mobility (μ_0) of 5 cm²/Vs and a gate-overdrive voltage ($|V_{GS}-V_{DS}|$) of 2 V.

Table 51 Literature review of voltage-normalized transit in equencies reported for organic 11 15.							
Reference	Substrate	Device	Voltage (V)	f_T/V (MHz V ⁻¹)			
Brown, Science, vol. 270, p. 972, 1995	Rigid	RO*	20	$1.25 \cdot 10^{-4}$			
Crone, J. Appl. Phys., vol. 89, p. 5125, 2001	Rigid	RO*	100	$5 \cdot 10^{-4}$			
Baude, Appl. Phys. Lett., vol. 82, p. 3964, 2003	Rigid	RO*	50	$6.67 \cdot 10^{-4}$			
Sheraw, Int'l Electr. Dev. Meeting 2000	Flexible	RO*	20	0.00125			
Fix, Appl. Phys. Lett., vol. 81, p. 1735, 2002	Flexible	RO*	80	0.0092			
Wagner, Appl. Phys. Lett., vol. 89, p. 243515, 2006	Rigid	RO*	10	0.2			
Heremans, Int'l Electr. Dev. Meeting 2009	Flexible	RO*	20	0.2			
Zschieschang, Org. Electronics, vol. 14, p. 1516, 2013	Flexible	RO*	4	0.42			
Kitamura, Appl. Phys. Lett., vol. 95, p. 023503, 2009	Rigid	TFT ^a	25	0.8			
Kitamura, Jpn. J. Appl. Phys., vol. 50, p. 01BC01, 2011	Rigid	TFT ^a	25	1.11			
Zaki, Org. Electronics, vol. 14, p. 1318, 2013	Rigid	TFT ^b	3	1.37			
Nakayama, Adv. Mater. Interfaces, vol. 1, p. 1300124, 2014	Rigid	TFT ^a	10	1.9			
Yamamura, Sci. Adv., vol. 4, p. eaao5758, 2018	Rigid	TFT ^a	10	2			
Perinot, Adv. Sci., vol. 6, p. 1801566, 2019	Flexible	TFT ^a	14	2.06			
Borchert, Int'l Electr. Dev. Meeting 2018	Flexible	TFT ^b	3	2.23			
Kheradmand-Boroujeni, Sci. Rep., vol. 8, p. 7643, 2018	Rigid	TFT ^c	8.6	4.65			
This work	Flexible	$\mathrm{TFT}^{\mathrm{b}}$	3	7			

Table S1| Literature review of voltage-normalized transit frequencies reported for organic TFTs.

^a Small-signal currents directly measured to evaluate f_{T} .

^b S-parameter measurement to evaluate f_{T} .

^c Pulsed-bias measurement circuit to evaluate f_{T} .

*In cases where the data were obtained from measurements on ring oscillators (RO), the equivalent frequency $f_{eq} = 1/(2\tau)$ is normalized to the supply voltage.

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