Embedded Dipole Self-Assembled Monolayers for Contact Resistance Tuning in p-Type and n-Type Organic Thin Film Transistors and Flexible Electronic Circuits

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Based on the powerful concept of embedded dipole self-assembled monolayers (SAMs), highly conductive interfacial layers are designed, which allow tuning the contact resistance of organic thin-film transistors over three orders of magnitude with minimum values well below 1 kΩ cm. This not only permits the realization of highly competitive p-type (pentacene-based) devices on rigid as well as flexible substrates, but also enables the realization of n-type (C60-based) transistors with comparable characteristics utilizing the same electrode material (Au). As prototypical examples for the high potential of the presented SAMs in more complex device structures, flexible organic inverters with static gains of 220 V/V and a 5-stage ring-oscillator operated below 4 V with a stage frequency in the range of the theoretically achievable maximum are fabricated. Employing a variety of complementary experimental and modeling techniques, it is shown that contact resistances are reduced by i) eliminating the injection barrier through a suitable dipole orientation, and by ii) boosting the transmission of charge carriers through a deliberate reduction of the SAM thickness. Notably, the embedding of the dipolar group into the backbones of the SAM-forming molecules allows exploiting their beneficial effects without modifying the growth of the active layer.

1. Introduction

Contact resistances hamper the development of organic thin-film transistors (OTFTs) with operation frequencies in the MHz–GHz range.[1,2] This is particularly true when realizing high-frequency operation by reducing the transistor channel length, as then the contact resistance ($R_c$) can easily dominate over the channel resistance diminishing the possibility to control the transistor current via the gate voltage.[2] Consequently, several methods have been suggested for reducing $R_c$ especially in bottom-contact bottom-gate OTFTs (BC OTFTs), in which the contacts are fabricated before depositing the semiconductor. Strategies for reducing $R_c$ comprise, for example, O2 plasma[3] or ozone treatment[4] of the electrodes, improved electrode fabrication procedures,[5,6] interfacial doping,[7,8] using chemically modified organic metal alloys,[9] and the application of self-assembled monolayers (SAMs).[5,6,8,10–12] In this way, employing a pentafluorobenzenethiol SAM, contact resistances as low as 0.29 kΩ cm have been realized at an operation voltage as high as 8 V.[13]

In fact, SAMs are beneficial for OTFTs in multiple ways:[8,14–15] They allow an adjustment of the energy level alignment between electrode and semiconductor,[16–18] they improve the OTFT reliability, they help increasing crystallinity and...
molecular packing of the organic semiconductor,\cite{10,19} they
serve as adhesion layer to prevent irregular contact edges and
voids,\cite{16} and they remove contaminants from the surface by
replacing them with covalently linked molecules.

To maximize the favorable properties of SAM-modified
contacts, several conditions ought to be met: i) Independent
of whether aliphatic\cite{6,15,20−27} or aromatic\cite{6,10,17,21,28−32} back-
bones are employed in the devices, it must be possible to
alter efficiently the chemical structures of the molecules such
that the level alignment at the electrode/semiconductor inter-
face can be adjusted in a straightforward manner. Ideally, the
chosen structural motifs should be flexible enough to provide
the possibility to realize monolayers that allow either elec-
tron or hole injection into suitably chosen semiconductors
from a single type of electrode. ii) The SAM structures used
to induce the desired changes in the substrate work-function
should not compromise the growth of the active organic semi-
conductor layer.\cite{19} Thus, systems in which level alignment
and semiconductor growth can be tuned independently are
particularly promising. iii) To maximize the injected currents,
carrier transport through the monolayers has to be as efficient
as possible; i.e., SAMs should by themselves be as transmis-

sive for electrons or holes as possible.\cite{33} iv) For a truly knowl-
edge-based design of SAM-modified electrodes, it is crucial to
combine an in-depth understanding of the SAM properties
with an extended characterization of the organic layers grown
on the SAMs, suitable modeling approaches both at the atom-
istic and at the device level, and a thorough investigation of
the ensuing device structures. v) To allow the realization of
more complex, multicomponent electronic circuits, it must
be possible to incorporate the SAM deposition onto the con-
tacts into a highly reproducible and simple OTFT fabrication
process.

All these aspects are realized in the present manuscript
with a versatile SAM platform, which meets the above require-
ments in an elegant and highly adaptive way. An outstanding
feature is that this SAM concept allows the straightforward
realization of both p-type and n-type transistors and circuits
using the same electrode material and geometry with mini-
mized contact resistances (on the order of 1 kΩ cm for devices
operated at ≈ 3 V). Thus, instead of multistep and error-prone
process routes for fabricating combinations of contact materi-
als, we simply need to immerse the gold electrodes in a
solution of the SAM-forming molecules with the appropriate
dipole orientation. This can be done without the need to
adapt the semiconductor growth conditions to the electrode
material. Moreover, due to the embedded-dipole approach,
detrimental effects of the SAM on the morphology of the
semiconductor deposited on top of the SAM are avoided. The
employed strategy increases the device-to-device reproduc-

ibility (especially on flexible substrates), improves yields, and
saves time and costs; these aspects are all inevitable prereq-

uities for the large area production of complex organic cir-
cuits. As prototypical examples of such circuits, we realized
flexible unipolar inverters with high static gain and 5-stage
ring-oscillators.

Our SAM platform is based on an aromatic backbone to min-
imize additional tunneling barriers caused by the interfacial
layer.\cite{11,34−38} In a first step, we used terphenyl-4-methanethiol
(TP1; TP referring to the terphenyl backbone and 1 denoting
one methylene spacer). To trigger work-function changes, a
polar 2,5-pyrimidine moiety is introduced into the backbone,
as shown in Figure 1a (first generation).\cite{37,38} Employing mixed
monolayers of these molecules, it is even possible to continu-
ously tune the work function of a Au substrate over a range of
0.9 eV.\cite{37} To further boost the beneficial effect of the SAMs, we
deliberately synthesized molecules derived from a biphenyl-
4-thiol (BP0) backbone that is shorter and has no methylene
spacer in the linker (see Figure 1a, “2nd generation”), thus
hoping to minimize the detrimental tunneling barrier due to
the SAM.

2. Results and Discussion

2.1. SAM Characterization

To assess the suitability of the pyrimidine-substituted SAMs
for engineering Au surfaces serving as electrodes in OTFT
contacts, we first grew SAMs on plasma-cleaned Au layers
deposited on glass-based OTFT substrates according to a pro-
cess described in the Experimental Section and illustrated
in Figure S1 in the Supporting Information. The embedded
dipoles in the SAMs pointing downward (upward) cause the
work function (W) to increase (decrease) with respect to the
dipole-free reference SAMs. For both generations of SAMs,
the covered work-function range amounts to at least ≈0.9 eV, as
shown by Kelvin-probe (KP) measurements, W_{F,KP} (Figure 1b),
and via the secondary electron cutoffs W_{ESCA} in the Ultra-

violet photoelectron spectroscopy (UPS) spectra (Table 1;
Figure S3 in the Supporting Information). To trace the origin
of that shift, we calculated the electrostatic energy in the SAM
employing dispersion-corrected density-functional theory using
the repeated slab approach (with the Perdew, Burke, Ernzerhof
(PBE) exchange functional).\cite{39} The results for BP0-up and BP0-
down, shown in Figure 1c, reveal that the potential drop pri-
marily occurs within the layer in the spatial vicinity of the N
atoms. Notably, the energy has become essentially constant at
the top of the SAM.

Grazing incidence X-ray diffraction (GIXD) measurements
show that pentacene grows on SAM-treated electrodes in
crystallites consisting of upright standing molecules. These
crystallites belong to two polymorphs, the thin film phase, and
the Campbell phase.\cite{40,41} The structures are essentially
the same on all surfaces; small deviations are found only for
the growth on the TP1 surface (see Figure S4 of the Supporting
Information). On the Al2O3 surface used as gate dielectric in
the devices (see below), the GIXD measurements show the
presence of only the pentacene thin film phase.

To assess the film morphology, we performed atomic force
microscopy (AFM) experiments and to relate the morphology
to the properties of the underlying SAM-covered electrodes,
we conducted contact angle (CA) measurements. Consistent with
all SAMs having a surface energy higher than that of pentacene
(γ_{pentacene} = 42.1 mJ m\(^{-2}\))\cite{42} (cf. Table 1), we observe pronounced
Stranski–Krastanov-type island growth (Figure 2). The mor-
phologies of the pentacene films on top of all 1st generation
SAMs are very similar (see Figure 2a). They are fine-grained
with nucleation densities $N$ between 20.3 and 23.8 grains $\mu m^{-2}$. This is consistent with similar values of the surface energy, $46.4 \text{ mJ m}^{-2} \leq \gamma \leq 50 \text{ mJ m}^{-2}$ for all three SAMs.

For the shorter and purely aromatic 2nd generation SAMs, we observe the following differences compared to the first-generation systems: The RMS surface roughness values of BP0-based SAMs are significantly smaller than those of TP1-based SAMs and the surface energies of BP0 and BP0-down SAMs are lower. Both aspects lead to a change of the semiconductor morphology.[42–46] Consequently, on the 2nd generation SAM treated Au surfaces, we observe larger average sizes of pentacene grains (see Figure 2b) along with lower nucleation densities $N$. On average, the morphologies of the pentacene layers are very similar to the ones observed on blank, plasma-cleaned Au-layers but with higher uniformity (see Table S1 and Figure S5d in the Supporting Information). The decrease in $N$ is more pronounced for BP0-down and BP0 SAMs ($N \approx 7.3–7.9$ grains $\mu m^{-2}$; Table 1). For growth on BP0-up SAMs, the nucleation

Table 1. Work function values, surface properties, and nucleation density $N$ of pentacene on different SAM treated Au layers.

| Gen | SAM         | $W_{(KP)}$ [eV] | $W_{(UPS)}$ [eV] | $\gamma_D$ [mJ m$^{-2}$] | $\gamma_P$ [mJ m$^{-2}$] | $\gamma_{tot}$ [mJ m$^{-2}$] | RMS [nm] | $N$ [grains $\mu m^{-2}$] |
|-----|-------------|----------------|------------------|-------------------------|-------------------------|-----------------------------|----------|--------------------------|
| 1st | TP1-down    | 4.87 (0.1)     | 4.75 (0.1)       | 49.9 (0.6)              | 47.8 (0.4)              | 2.10 (0.2)                  | 1.90 (0.1) | 23.2 (2.3)               |
|     | TP1         | 4.45 (0.1)     | 4.30 (0.1)       | 46.4 (0.4)              | 46.0 (0.3)              | 0.40 (0.1)                  | 1.70 (0.1) | 23.8 (4.6)               |
|     | TP1-up      | 4.01 (0.1)     | 3.90 (0.1)       | 50.0 (0.6)              | 47.8 (0.4)              | 2.20 (0.2)                  | 1.85 (0.1) | 20.3 (3.3)               |
| 2nd | BP0-down    | 4.83 (0.1)     | 4.95 (0.1)       | 44.2 (0.6)              | 42.6 (0.5)              | 1.60 (0.1)                  | 1.60 (0.1) | 7.3 (0.7)                |
|     | BP0         | 4.46 (0.1)     | 4.30 (0.1)       | 43.7 (0.5)              | 42.1 (0.4)              | 1.60 (0.1)                  | 1.40 (0.1) | 7.9 (0.8)                |
|     | BP0-up      | 4.07 (0.1)     | 3.75 (0.1)       | 57.3 (0.8)              | 46.6 (0.6)              | 10.7 (0.2)                  | 1.60 (0.1) | 10.9 (2.8)               |

$^4W_{(KP)}$ and $W_{(UPS)}$ refer to work functions determined by Kelvin probe and from secondary electron cut-offs of UPS-spectra. The measurements have been performed on SAMs prepared on substrates equivalent to those used for device fabrication; $^4\gamma$ is the total surface energy extracted from contact angle measurements with $\gamma_D$ and $\gamma_P$ being its dispersive and polar components; $^4$RMS is the root mean square of the surface roughness, and $N$ the grain density. The listed numbers correspond to the mean values over four experiments for each monolayer with the respective standard deviations cited in brackets.
density ($N \approx 10.9 \text{ grains } \mu m^{-2}$) is somewhat higher, but still far below that one on the TP1-based SAMs. We attribute the higher $N$-value to the distinctly higher polar component of the surface energy originating from pyrimidine as the terminal ring of the BP0-up molecule.

2.2. OTFTs with Embedded Dipole SAM-Treated Au Electrodes

Knowing the key electronic, structural, and morphological properties of the studied layers, the next step is to incorporate them into the bottom gate, bottom contact OTFT architecture shown in Figure 3a. This architecture features an 18 nm ultrathin Al₂O₃ gate dielectric that allows for operation voltages below 3 V due to its large gate capacitance of 440 nF cm$^{-2}$ and its remarkable density (gate leakage currents below 150 pA, see Table S3 in the Supporting Information). A thiol-terminated phosphonic acid SAM (PA-SAM) immobilized on the Al₂O₃ substrate serves as an adhesion layer for the Au electrodes patterned by photolithography. Channel lengths were varied between 7.5 and 51 µm and channel widths between 1 and 4 mm. At each channel length, several devices were produced (e.g., 5–10 transistors with $L \approx 7.5 \mu m$ for dipole-down SAM treated electrodes). For the extraction of the contact resistance, several transmission line method (TLM) sets were fabricated (e.g., 3–5 TLM-sets of transistors with dipole-down SAM treatment), each made of 4–6 transistors with different channel lengths. The number of considered devices is specified in detail in the respective table captions. More information on the fabrication process and characterization of the devices can be found in the Experimental Section.

Typical $I_d(V_{DS})$-electrical output characteristics for bottom contact OTFTs with SAM-modified source and drain electrodes for channel lengths of $L = 7.5 \mu m$ are shown in Figure 3b. The magnitude of the current changes by a factor of $\approx 20$ going from down to up SAM modified OTFTs. This is consistently observed for applying the 1st (upper panels) and the 2nd generation SAMs (lower panels). Since within each SAM generation, the pentacene morphology on the electrodes is very similar irrespective of the direction of the embedded dipole moment, we attribute these differences primarily to dipole-induced changes in the hole-injection barriers (HIB). The latter can be estimated from the (SAM-modified) work-function of the electrode ($W_f$) and the ionization potential of upright standing pentacene molecules ($I_{p,pentacene}$)[47,48] as $\text{HIB} = I_{p,pentacene} - W_f$, considering that no significant interface dipoles[49,50] are expected in the contact region between the SAM and the organic semiconductor. In all cases in which $I_{p,pentacene} < W_f$, we set HIB = 0 due to Fermi level pinning (see Table S3 in the Supporting Information). Transistor parameters for the reference samples with $H_2$ and $O_2$-plasma cleaned Au electrodes and no SAM treatment are listed in Table S2 in the Supporting Information; exemplary $I(V)$ characteristics are displayed in Figure S5a–c (Supporting Information).

Figure 2. AFM height-images of 40 nm thin pentacene films on the a) 1st and b) 2nd generation SAMs on Au.
For SAMs containing no or upward-pointing dipoles, the sizable values of HIB between 0.35 eV and ≈ 0.8 eV give rise to large $R_c$ values. These also cause the S-shape in the low voltage region of the measured output curves.[51] By contrast, the SAMs with downward pointing dipole moment (TP1-down, BP0-down) and vanishing HIBs (see Table S3, Supporting Information) develop a perfectly linear $I_D(V_{DS})$ characteristics in the respective region.

Further insight in the influence of the dipole-SAM treatment on the device parameters is provided by the transfer curves (e.g., Figure S6a,b in the Supporting Information). Since the ON current reaches a maximum in devices with dipole-down SAMs on Au (TP1-down and BP0-down), whereas the OFF current is virtually constant (see Table S3, Supporting Information), the ON/OFF current ratio increases continuously from dipole-up over no dipole to dipole-down SAM treated Au electrodes with best values for OTFTs with the shorter BP0-down SAMs on Au (see Table S4, Supporting Information). The dipole-down SAMs also have the smallest sub-threshold swing $S$, and the lowest positive onset voltage $V_{ON}$, which indicates a smaller contribution from the voltage across the Schottky barrier at the charge injecting interface (see Equation (S4) in the Supporting Information).

To quantify $R_c$, we analyzed all sets of output and transfer curves in two distinct ways. For a comparison with the bulk of literature, we first determine $R_c$ by systematically varying the channel length and employing the TLM described in the Supporting Information (see also Figures S7 and S8 in the Supporting Information).[52,53] As an alternative, to explicitly discriminate between ohmic (linear) and nonohmic (nonlinear) resistance contributions[53] (the latter arising from the injection barrier between electrode surface and semiconductor), we determine $R_c$ with a newly developed fitting approach (FA) described in detail in the Supporting Information (see also Figure S9, Supporting Information). In short, inspired by the suggestion of Fischer et al.,[54] we simultaneously fit the output and transfer curves for different operation conditions to an equivalent circuit model combining an ideal transistor with a gate-controlled Schottky diode. The characteristics reconstructed from the fitted model parameters fully reproduce the measured curves, in particular in terms of their pronounced S-shapes at large HIB (Figure 3b, magenta lines).

Figure 3. a) Schematic 3D representation of the bottom gate, bottom contact OTFTs with pyrimidine-containing 2nd generation molecules (here BP0-up) assembled on the Au electrode. The red layer underneath the photolithography patterned source–drain electrodes represents the adhesion layer (phosphonic acid SAM). b) Typical output characteristics ($I_D(V_{DS})$) for $L = 7.5 \mu m$ obtained for contact treatment with SAMs composed of the 1st (upper panels) and 2nd generation molecules (lower panels). The measured data points are plotted in black, while the results from the fitting analysis (described in the main text and in the Supporting Information) are drawn in magenta.
2nd generation SAMs (BP0-based, filled symbols) systematically exceed the ones for the 1st generation SAMs (TP1-based, open symbols). Mobility values of transistor parameters and extracted contact resistance values vary by more than two orders of magnitude within each generation of SAMs. The full set of transistor parameters and extracted contact resistance values are associated with up SAMs (light blue shaded region in panel (f) cannot be compared in terms of generations, because the transport is channel-resistance-dominated, while the unshaded regions indicate contact-resistance-dominated transport. In (f), the \( R_c \) values, derived from the fitting analysis of SAM-modified transistors of a transmission line series, are plotted as a function of the fitted contact resistance \( R_{c,FA} \). The shaded regions indicate that transport is channel-resistance-dominated, while the unshaded regions indicate contact-resistance-dominated transport. In all graphs, open and filled symbols correspond to 1st (TP1-based) and 2nd generation (BP0-based) SAMs, respectively.

As shown in Figure 4a,b for the two analysis approaches, the obtained channel-width normalized contact resistances \( R_c \) for different dipole orientations (and HIBs) vary by more than two orders of magnitude within each generation of SAMs. The full set of transistor parameters and extracted contact resistance values are found in Table S4 in the Supporting Information. Notably, for the dipole-down SAMs on Au (TP1-down, BP0-down) the contact resistances are purely ohmic and are clearly below 10 kΩ cm (cf. Table S4, Supporting Information). For BP0-down, they even fall as low as 0.58 kΩ cm (best value, see Table 2). Much larger

Table 2. Critical transistor parameters and contact resistance values determined by TLM and FA for OTFTs with different SAM-treated S/D Au-electrodes on glass and on flexible polycarbonate (PC)-substrates.

| Gen. | Semiconductor/substrate | SAM | \( \mu_{c,FA} \) \([cm^2 V^{-1} s^{-1}]\) | \( \mu_{c,TLM} \) \([cm^2 V^{-1} s^{-1}]\) | \( R_{c,FA} \) \([kΩ cm]\) | \( R_{c,TLM} \) \([kΩ cm]\) |
|------|-------------------------|-----|---------------------------------|---------------------------------|-----------------|-----------------|
| 1st  | Pentacene/glass         | TP1-down | 0.1 (0.05) | 0.06 (0.03) | 0.09 | 7.5 (5.6–9.7) | 5.5 |
| 2nd  | Pentacene/glass         | BP0-down | 0.18 (0.04) | 0.17 (0.05) | 0.21 | 0.85 (0.58–1.3) | 2.4 |
|      | Pentacene/PC            | BP0-up | 0.20 (0.03) | 0.17 (0.04) | 0.15 (0.03) | 2.5 (1.7–3.5) | 1.6 (0.4) |
|      | C_{60}/PC               | BP0-up | 0.23 (0.04) | 0.13 (0.03) | 0.09 (0.04) | 4.6 (3.5–5.6) | 3.5 (3.0) |

\( \mu_{c,FA} \) and \( \mu_{c,TLM} \) are the charge carrier mobilities in the saturation and linear regime at \( V_{GS} = V_{ON} = -3 V \); for pentacene OTFTs the listed \( \mu_{c,FA} \) and \( \mu_{c,TLM} \) values (standard deviation given in brackets) are averaged over 5 transistors on glass and 10 transistors on PC-substrates and for the \( C_{60} \) OTFTs over 10 devices, all devices have channel lengths in the range \( L = 5–7.5 \) \( \mu \)m; \( \mu_{c,FA} \) is the \( R_c \)-corrected equivalent mobility in the linear regime at \( V_{GS} = V_{ON} = -3 V \) (the value for glass corresponds to the analysis shown in Figure 4b); \( R_{c,TLM} \) is the width-normalized contact resistance calculated in the linear operation regime (at \( V_{GS} = V_{ON} = -3 V \)) obtained by the TLM from OTFTs with four different channel lengths (7.5–51 \( \mu \)m) on a glass substrate and with six different channel lengths (3.5 \( \mu \)m \( \leq L \leq 52 \) \( \mu \)m) on a flexible PC-substrate; on glass substrates the listed \( R_{c,TLM} \) values are averaged over three TLM-sets, whereas on the PC substrates there are five TLM-sets for pentacene and two for \( C_{60} \), the respective min.–max. values are indicated in brackets; \( R_{c,FA} \) is the normalized contact resistance from FA. On glass, \( R_{c,FA} \) is obtained for the OTFT with \( L = 7.5 \) \( \mu \)m; on the PC substrate, the value of \( R_{c,FA} \) corresponds to the average over all OTFTs with 4.5 \( \mu \)m < \( L < 10 \) \( \mu \)m.
$R_c W$ values are obtained for dipole-free SAMs (TP1, BP0; 100 kΩ cm ≤ $R_c W$ ≤ 1000 kΩ cm) and for the dipole-up systems (TP1-up, BP0-up; $R_c W = 1000$ kΩ cm). The employed fitting routine shows that these high contact resistances are dominated by contributions that are nonlinear in $V_C$ and, thus, arise from injection processes across a barrier (cf. Table S4, Supporting Information). This nonlinear contribution is, in fact, also responsible for the pronounced S-shapes in the output curves discussed above.[35] The $R_c W$ values for devices without any SAM treatment (see Table S2, Supporting Information) are similar on average to those of TP1-down treated OTFTs but with a much larger spread.

When comparing transistors containing SAMs with different backbone structures, the $R_c$-values for 2nd generation, BP0-derived SAMs are consistently lower compared to 1st generation, TP1-derived ones. For the dipole-down SAMs, this difference reaches nearly an order of magnitude (see Table 2). This might seem insofar surprising, as based on the measured work functions (see Figure 1b), there should be essentially identical hole-injection barriers into the pentacene layer for a given dipole orientation. The more favorable morphology of the pentacene layers on BP0-based SAMs should also primarily result in a slight improvement of the carrier mobility (see below). Therefore, we attribute the better performance of OTFTs treated with the 2nd generation molecules predominantly to a higher conductance of the BP0-based SAMs due to the shorter length of the conjugated backbone and the absence of the methylene linker.[33] Indeed, conductivity measurements through the SAMs on Au using an EGIn droplet as counter electrode[36] (Figure 4c) show that for a monolayer junction the currents through 2nd generation up and down molecules (blue) are 1–2 orders of magnitude higher compared to the 1st generation ones (red). A more detailed discussion of the energetic alignment between the states in the SAMs and the Fermi-level of the substrate can be found in Figures S10 and S11 (Supporting Information).

Contact resistances also determine the extent to which the effective charge carrier mobility, $\mu$, extracted from an OTFT characterises, represents the actual property of the organic semiconductor.[57] Typically, mobility values in the saturation regime, $\mu_{sat}$, and in the linear regime, $\mu_{lin}$, are extracted from the transconductance $dI_{DS}/dV_{GS}$.[37] To remove the influence of the contact, we determined the equivalent mobility in the transistor channel, $\mu_{eqv,F}$ for negligible drain-source bias from the abovementioned fitting approach (see Table S4, Supporting Information). Figure 4d–f shows mobility values for different SAM-modified OTFTs as a function of the width-normalized contact resistance; each panel collects all values regardless of the different channel lengths and SAM generations. The effective mobility in the saturation regime (Figure 4d) is independent of the channel length only for $R_c W \leq 10$ kΩ cm (shaded region, channel-resistance dominated transport). For higher values of $R_c W$, $\mu_{sat}$ undergoes a clear transition to a contact-resistance dominated transport regime with a strongly $L$-dependent, effective mobility. That is, in that regime, the extracted value of $\mu_{sat}$ is primarily determined by the contact resistance and is no longer a direct property of the active material. The linear mobility is even more affected by the contact resistance, with $L$-independent values for $\mu_{lin}$ only for $R_c W = 1$ kΩ cm, i.e., only for BP0-down SAMs/Au (shaded region in Figure 4e). In stark contrast, the $R_c$-corrected mobility $\mu_{eqv,F}$ (Figure 4f) does not systematically depend on the channel length or on $R_c W$ (respectively, the dipole orientation). Rather, $\mu_{eqv,F}$ of the 2nd generation (filled symbols) typically surpasses the $\mu_{eqv,F}$ of the 1st generation. Values in the shaded region are excluded from this comparison because of the lower quality of the fit in the presence of dominating nonlinear $R_c$ contributions. The remnant spread of the values stems from device to device fluctuations between different channel lengths.

Although it does not directly relate to the discussion of the influence of $R_c$ on $\mu$, it has to be noted that even our devices with ultralow $R_c$ have not reached the best mobility values reported for staggered pentacene OTFTs on high-k dielectrics; our mobilities are smaller by a factor of approximately three for Al$_2$O$_3$[58] or approximately six for ZrO$_2$-PaMS nanocomposites.[59] Despite very low evaporation rates, our pentacene layers presumably contain more grain boundaries and thus a smaller intrinsic mobility due to two reasons: First, the anodized Al$_2$O$_3$ surface, that emerged after the photolithographic process and the plasma cleaning, is not pristine, because we had to omit a passivating and smoothing SAM treatment of the dielectric to avoid any cross-influence on the pyrimidine SAMs. In order to allow for a clear assignment of the contact resistance tuning to the type of embedded dipole SAMs on the contacts, we, thus, deliberately accepted a dielectric surface that is good but not perfect for OSC growth with small grain boundaries. Second, despite the SAM treatment of the contacts, we cannot exclude that our coplanar setup causes a less homogeneous film growth in the contact-channel transition region.

### 2.3. p-Type and n-Type OTFTs with Embedded Dipole SAM-Treated Au Electrodes on Flexible Substrates

The effective lack of injection barriers in OTFTs containing TP1-down or BP0-down SAMs leads also to reduced spreads in the OTFT parameters and to the smallest subthreshold swings in the transfer characteristics of all devices studied here (Table S3 in the Supporting Information). This makes such devices particularly promising for applications in more complex electronic circuits. Realizing, for example, inverters and ring oscillators on mechanically flexible substrates would be particularly interesting, as it could significantly impact the area of flexible electronics (Figure 5a). Additionally, a possibility to fabricate n-type devices on flexible substrates without changing the electrode material would be highly intriguing. Consequently, the next logical step is to test the transferability of our concept for contact engineering to devices on flexible substrates and to n-type organic semiconductors (in both instances varying channel lengths between 3.5 and 52 μm).

BP0-down SAM modified pentacene OTFTs ($L = 7.5$ μm) fabricated on a flexible polycarbonate (PC) substrate yield output (Figure 5b) and transfer characteristics (Figure S12a, Supporting Information) that favorably compare to the OTFTs on glass discussed above.

Table 2 compares the short channel transistors ($L = 5–7.5$ μm) with optimum SAM treatment on glass and on PC substrates regarding mobility and contact resistance values. Notably the mobilities ($\approx 0.2$ cm$^2$ V$^{-1}$ s$^{-1}$) and the ON/OFF ratios
≈10^6) are essentially identical (for the full set of parameters, see Table S5 in the Supporting Information). Also the \( R_c \cdot W \) values amounting to \( ≈2.5\ \text{kΩ cm} \) are essentially the same as for the equivalent glass-based OTFTs when employing the more sophisticated fitting analysis. We believe that this competitive device performance on the flexible substrates is due to the excellent surface quality of our PC substrate, which is exceptional in terms of surface roughness (0.9 nm) and process stability owing to a special hard coating layer. Finally, it has to be noted that mechanical stress tests (bending or strain tests) for pyrimidine-SAM treated p-type OTFTs are ongoing and will be reported in the near future.

For fabricating n-type devices, BP0-up SAMs should be particularly promising considering that they efficiently lower the electron injection barrier (EIB). For example, for C60-based OTFTs, a barrier of \( E_{IB} = W_{F,KP} - E_{A_{C60}} \approx 0.15\ \text{eV} \) (for \( E_{A_{C60}} \approx 3.92\ \text{eV} \)) can be anticipated. The high application potential of the dipole-up SAMs for contact resistance reduction is fully confirmed by the performance of the C60 OTFTs that we fabricated on flexible PC-substrates. The corresponding output characteristics for OTFTs containing 2nd generation SAMs with \( L = 7.5\ \mu\text{m} \) are shown in Figure 5c, the transfer characteristics in Figure S12b (Supporting Information). They reveal a clear n-type behavior for all three SAMs. However, only for the BP0-up modified electrodes, \( I_D(V_{DS}) \) curves without S-shape are observed. In that case, we observe an average contact resistance as low as \( R_{c,TLM} \cdot W = 4.6\ \text{kΩ cm} \) and a corrected mobility of \( \mu_{eqv,FA} = 0.09\ \text{cm}^2\ \text{V}^{-1}\ \text{s}^{-1} \) (see Table 2), which are both in the same range as for the BP0-down treated, pentacene-based p-type OTFTs on the PC-substrate.

### 2.4. Flexible Inverters and Ring-Oscillators Containing Embedded-Dipole SAMs

The excellent performance and the small device-to-device variation (see Table S5, Supporting Information) observed for the flexible BP0-down modified p-type OTFTs qualify these transistors for integration into flexible organic circuits. Accordingly, as representative examples, we fabricated unipolar inverters and 5-stage organic ring-oscillators on a flexible 1 in. × 1 in. polycarbonate substrate.

Typical voltage transfer characteristics (VTC) of a zero-\( V_{GS} \) inverter for supply voltages of \( V_{DD} = 2, 3, \) and 4 V are shown in Figure 6a. Up to an operation voltage of \( V_{DD} = 4\ \text{V} \), the inverter exhibits a high static gain of 220 V/V, a very sharp transition regime (\( \Delta V_{in} < 140\ \text{mV} \)) and full rail-to-rail swing. Figure 6b also demonstrates the remarkably large noise margin of 0.82 V and the symmetric transition close to \( V_{DD}/2 \) for forward and reverse voltage sweeps.\(^{[61]}\)

A photograph of a flexible 5-stage ring-oscillator made of such inverters and the measured output signal at \( V_{DD} = 4\ \text{V} \) are displayed in Figure 6c. The output signals of two oscillators on the same sample are displayed in Figure S13 (Supporting Information). They show output frequencies in the range of 0.35–0.56 kHz (\( V_{DD} = 4\ \text{V} \)) corresponding to a minimum stage delay \( \tau_{min} \approx 148–236\ \mu\text{s} \) and thus a stage frequency of \( f_S \approx 3.4–2.1\ \text{kHz} \).\(^{[62]}\) This value is encouraging, considering the calculated maximum transfer frequency, \( f_T \), of 11 kHz for a bottom contact pentacene-based BP0-down modified OTFT. The inability to reach the theoretical maximum is not the
consequence of a contact resistance, but is rather the consequence of a parasitic capacitance contribution from the stage interconnections and due to the load transistor being operated just above the threshold. A detailed discussion of the performance of the inverters and ring oscillators and their transfer frequency is found in the Supporting Information.

3. Conclusion

In summary, in this work we show how the new concept of embedded-dipole SAMs can be adapted to tune the contact resistance in bottom contact, bottom gate organic thin-film transistors by three orders of magnitude. This tuning is achieved by switching the orientation of a pyrimidine ring incorporated in the aromatic backbones of the SAM-forming molecules, which allows modifying the work function of the electrodes over a range of 0.9 eV (verified by Kelvin probe and UPS). Quantum-mechanical simulations show that the underlying shift in the electrostatic energy occurs within the self-assembled monolayer. The concept of embedding the dipolar elements into the molecular backbones ensures that switching the dipole orientation neither impacts the crystal structure of the organic layers (confirmed by X-ray diffraction), nor their morphology (as seen in atomic force microscopy). Varying the length of the aromatic systems and including/omitting a methyl spacer between the backbones and the thiolate docking group, we find that the reduced SAM-tunneling barrier in shorter and fully aromatic backbones allows modifying the work function of the electrodes over a range of 0.9 eV (verified by Kelvin probe and UPS). The best-performing p-type, pentacene devices operated at 3 V show width-normalized contact resistances as low as \( R_{cW} = 0.58 \, \text{k}\Omega \text{ cm on glass and 1.7 k}\Omega \text{ cm on plastic substrates as determined via the transfer-line method. A particularly intriguing aspect of the SAMs presented here is that by switching the dipole moment within the backbone, they also allow the realization of n-type, C_{60} based transistors on flexible substrates using photolithography patterned gold electrodes. Notably, the contact resistances and mobilities of the ensuing devices are very similar to those of the best p-type transistors.

To highlight the potential of SAMs with embedded dipoles for more complex device applications, we also fabricated functional 5-stage organic ring-oscillators on flexible substrates that were based on very well performing unipolar inverters (gain of \( \approx 220 \, \text{V/V} \) at 4 V) and operated below 4 V.

4. Experimental Section

While the synthesis of the TP1-derived molecule is described in ref. [38], details regarding the synthesis of the BPO-based molecules will be published elsewhere.

**Preparation of SAMs:** The SAMs were prepared by immersion of the freshly prepared substrates (cleaned “photopatterned” Au layers) into 100–500 \( \times \) 10^{-6} \, \text{mL} \, \text{ethanol} (\geq 99.9\%, HPLC grade, VWR) solutions of pyrimidine-substituted thiols and references molecules for 18 h. After immersion, the samples were carefully rinsed with pure solvent and blown dry with nitrogen.

**Tunneling Current Measurements across SAMs with Eutectic Ga–In (EGaIn) Top-Contacts:** Electrical conductance measurements were performed with a custom-made two-terminal tunneling junction setup, based on the Keithley 2635A source meter[61]. The gold substrate and a sharp tip of eutectic GaIn (EGaIn) served as the bottom and top electrodes, respectively.[56] Tunneling junctions were formed by contacting grounded SAM/Au samples with the EGaIn tips and applying a potential. The voltage was varied between \(-0.6 \) and \(+0.6 \, \text{V} \) in 0.05 V steps. At least 10 I–V curves measured at several different places were recorded for each sample; average values were calculated.

**Work Function Determination by UPS and KP:** UPS experiments were performed in a multiprobe surface analysis ultrahigh vacuum (UHV) system from Omicron Nanotechnology with a base pressure of 1 \( \times \) 10^{-10} \text{mbar}. The spectra were recorded using a HPS 1311 vacuum ultraviolet source (incident photon energy of 21.22 eV, He I line) and an EA 125 hemispherical analyzer with a pass energy of 1 eV and an energy resolution of 80 meV. The secondary cut-off was recorded while the sample was biased at \(-5 \, \text{V} \) (relative to the analyzer). The absolute error in the determination of \( W_f \) is estimated to be \( \pm 100 \, \text{meV} \).
to the UPS experiments, work function measurements were also carried out using a UHV Kelvin Probe 2001 system (KP technology Ltd., UK). The pressure in the UHV chamber was \(10^{-9}\) mbar. Freshly sputtered gold and hexadecane SAMs on Au(111) were used as references with the work function values set at 5.2\(^{[64]}\) and 4.32 eV\(^{[43]}\), respectively.

**Surface Analysis—** H\(_2\) and O\(_2\) Plasma-Cleaned Au and Al\(_2\)O\(_3\): X-ray photoelectron spectroscopy was used to verify the quality of the H\(_2\) and O\(_2\)-plasma cleaning step prior to SAM coating. The measurements were performed in the UHV system mentioned above using a DAR 400 X-ray source (Al K\(_\alpha\) radiation \(h\nu = 1486.7\) eV), an XM 500 quartz crystal monochromator (energy width 0.15 eV), and a take-off angle of 90\(^\circ\). The pass energy of the hemispherical analyzer for the detailed element spectra was set to 20 eV resulting in a total energy resolution of 0.5 eV.

**Characterization of Pentacene Thin Film Morphology with AFM, and GIXD/XRD:** The surface roughness of the SAM-treated and non-treated Au layers was extracted from atomic force microscopy (VEECO Dimension 3100 AFM) topography images. AFM was also used to investigate the morphology of the organic semiconductor (pentacene). AFM images were analyzed using the free WSxM software from Nanotec Electronica.\(^{[41]}\)

Grazing incidence X-ray diffraction experiments were performed at the XRD1 beamline at the synchrotron ELETTRA, Trieste (Italy). Radiation with a wavelength of 1.4 Å was chosen. An incidence angle of 0.2\(^\circ\) was chosen for the primary beam. The diffraction pattern was taken with a Pilatus 2 M detector from Dectris (Switzerland) by integrating for 0.2 s. Data acquisition took place at a fixed sample position. Calibration of the detector in terms of detector–sample distance and the associated tilt angles was performed by using a LaB\(_6\) reference material filled in a capillary. Data processing was performed by using the in-house developed software GIDV; data from these images are merged and transferred from pixel space to reciprocal space using standard procedures.

**Fabrication Process of p-Type OTFTs on Glass:** Organic thin film transistors were fabricated in a bottom-gate, bottom-contact architecture according to the setup illustrated in Figure 3a. In Figure S1a (Supporting Information), the process flow is shown schematically.

The aluminum gate electrode was deposited by thermal evaporation of a 50 nm thick aluminum layer through a shadow mask at a rate of 1 nm s\(^{-1}\), followed by anodization of the aluminum to create an 18 nm thick aluminum oxide. A detailed description of the anodization process can be found in ref. \(^{[68]}\).

Prior to the thermal evaporation of a 50 nm Au layer, a SAM with a phosphonic acid anchor group and a thiol tail group (12-mercaptopododecylphosphonic acid, Sigma-Aldrich) was applied as adhesion layer. The source/drain electrodes were formed by photolithography employing the following steps: after deposition of a 50 nm Au layer, a photoresist (AZ1505, MicroChemicals) was applied by spin-coating, followed by a brief baking step. After UV illumination (\(λ = 365\) nm, \(E = 50\) mJ cm\(^{-2}\)) through a photomask (Compugraphics), the UV-exposed areas were easily removed by dipping the sample in a developer solution (AZ Developer from MicroChemicals). The photopatterned source–drain electrodes were briefly plasma (H\(_2\) and O\(_2\); applied successively) cleaned and treated with pyrimidine-containing SAMs as described in sub-section “Preparation of SAMs.” Finally, a 40 nm thick layer of pentacene for the p-type transistors, unipolar inverters, and ring-oscillators or a 60 nm thick C\(_60\) (from Sigma-Aldrich) layer for the n-type transistors was deposited by thermal evaporation through a shadow mask. Very low evaporation rates of 0.1 nm min\(^{-1}\) for the first 5 nm and 1.2 nm min\(^{-1}\) for the remaining 35 nm were used for the pentacene films and a constant rate of 0.3 Å sec\(^{-1}\) for the C\(_60\) films. After production, all devices on PC-substrates were protected from light and stored under argon atmosphere in a glovebox. The channel length of the OTFTs varied between 3.5 and 52 \(µ\)m and the channel width was fixed at 1 mm. The channel length of the diode-load and zero-V\(_{CS}\) ring oscillators was 7.5 \(µ\)m.

**Determination of Surface Energy:** Contact angle measurements with ultrapure water and diiodomethane were used to determine the surface energy with a KRUSS DSA 100 ContactAngle Measuring System. The surface energy was calculated via the Owens–Wendt–Rabel–Kaelble method, using contact angles of different liquids with known disperse and polar fractions of the surface tension.\(^{[69]}\)

**Electrical Characterization:** Electrical measurements on OTFTs, inverters, and ring-oscillators were carried out in a glovebox under argon, using a parameter analyzer from MB-Technologies, a manual probe station from Suess, and a digital oscilloscope from Tektronix (TDS2024). From the \(I_0 (V_{GS})\) plot of the square root of the drain current as a function of the gate bias, the threshold voltage and the onset (or, more correctly, the gate voltage at which \(I_D\) becomes detectable) can be extracted (Figures S1a–S1c).
employing the PBE functional\(^{[29]}\) combined with the surface version\(^{[30]}\) of the Tkatchenko–Scheffler dispersion corrections\(^{[21]}\) for the Van der Waals interactions. The system was modeled using periodic boundary conditions within the so called repeated slab approach, inserting a vacuum region of at least 20 Å in the z direction and including a self-consistently calculated dipole correction\(^{[22]}\) between the slabs, to electrostatically and spatially decouple them. The metallic substrate was modeled with five Au layers, holding the three bottom ones fixed during all the calculations and turning off the dispersion corrections between the Au atoms. Two molecules were put in a (3 × 3 × 3) rect unit cell, whose dimensions in the x and y directions were defined accordingly to the calculated Au lattice constant. The docking groups were placed in fcc-hollow sites and both cofacial and herringbone arrangements were tested, with the latter being energetically more favorable. The systems were optimized using the FHI-aims default “tight” setting and a 9 × 5 × 1 k-points grid. The total energy criterion for the self-consistency cycle was set to 10\(^{-6}\) eV and the optimizations were performed until the maximum residual force component per atom was below 0.01 eV Å\(^{-1}\).

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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