Influence of the gate leakage current on the stability of organic single-crystal field-effect transistors

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We investigate the effect of a small leakage current through the gate insulator on the stability of organic single-crystal field-effect transistors (FETs). We find that, irrespective of the specific organic molecule and dielectric used, leakage current flowing through the gate insulator results in an irreversible degradation of the single-crystal FET performance. This degradation occurs even when the leakage current is several orders of magnitude smaller than the source-drain current. The experimental data indicate that a stable operation requires the leakage current to be smaller than $10^{-9} \text{ A/cm}^2$. Our results also suggest that gate leakage currents may determine the lifetime of thin-film transistors used in applications.

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The study of organic semiconductor transistors aims at the development of organic electronics, for its advantages of being flexible, cheap and suitable for large-area production. So far, considerable research effort has been focused on the optimization of the organic layer to improve the performance of thin-film transistors. Much less attention has been devoted to other important device aspects, such as, for instance, the choice of the gate insulator.

Recent work has demonstrated that the gate insulator plays an important role in determining the device performance. In particular, it has been shown that in polymer as well as in single-crystal organic transistors the mobility of charge carriers is systematically larger the lower the dielectric constant of the gate insulator. This implies that the use of low-$\epsilon$ dielectrics will result in a higher device switching speed. In view of this result, it appears useful to investigate systematically how different properties of the gate insulator affect the behavior of organic transistors.

In this paper we use organic single-crystal FETs to investigate how a small leakage current through the gate insulator affects the stability of the device operation. Specifically, we have investigated the behavior of organic single-crystal FETs of different molecules (tetracene, rubrene, perylene) in combination with different dielectrics ($\text{Ta}_2\text{O}_5$, $\text{ZrO}_2$, and $\text{SiO}_2$). We find that, irrespective of the specific molecule and dielectric used, leakage current flowing through the gate insulator results in an irreversible degradation of the single-crystal FET operation. The degradation is not due to the electrical breakdown of the insulating layer and it also occurs when the leakage current is several orders of magnitude smaller than the source-drain current. From the experimental data, we conclude that a stable operation of organic single-crystal FETs requires the current leaking to the FET channel to be smaller than $10^{-9} \text{ A/cm}^2$.

The fabrication of the single-crystal FETs used in this work is based on electrostatic bonding of an organic single-crystal to a dielectric surface, with pre-fabricated source, drain and gate contacts. The details are essentially identical to what has been described in Ref. [2]. Whereas in Ref. [4] only thermally grown $\text{SiO}_2$ was used as gate insulator, here we have also used sputtered layers of $\text{Ta}_2\text{O}_5$ and $\text{ZrO}_2$ deposited in different ways. For both $\text{Ta}_2\text{O}_5$ and $\text{ZrO}_2$ we have investigated FETs in which the dielectric layers were sputtered from ceramic targets (hereafter referred to as "type I" oxides; see Ref. [10] for details). For $\text{Ta}_2\text{O}_5$, we have also investigated the behavior of FETs fabricated on layers sputtered from a metallic target, in the presence of oxygen in the sputtering gas, as described in Ref. [11] (hereafter referred to as "type II" $\text{Ta}_2\text{O}_5$). For all FETs discussed here the sputtered oxide layers were approximately 350 nm thick.

The electrical properties of all the different dielectric layers were characterized by capacitance and $I$-$V$ measurements (see [10]). From these measurements we obtain a dielectric constant $\epsilon = 25$ for $\text{Ta}_2\text{O}_5$ (both types) and 23 for $\text{ZrO}_2$, as expected. The breakdown field is comparable for all layers and typically equal to $1.5 \text{ MV/cm}$.

**FIG. 1:** Current-voltage characteristics of an organic single-crystal FET, with tetracene as the organic molecule and type I $\text{Ta}_2\text{O}_5$ as the gate insulator.
The anomalous behavior reproducibly exhibited by type I Ta$_2$O$_5$ and ZrO$_2$ FETs originates from irreversible device degradation. Specifically, we observe that, for every device studied, repeating the measurement of the $I_D$-$V_G$ curve systematically results in lower measured values of $I_D$ (see Fig. 2b). For those type I Ta$_2$O$_5$ transistors in which the increase in $V_G$ is sufficient to fully suppress the source-drain current (see Fig. 2b), no field-effect induced current is ever observed after the measurement, indicating that the degradation of the device is complete.

Inspection of the degraded transistors using an optical microscope does not reveal any visible change in the device. The bulk of the crystal, the dielectric layer, and the FET circuitry appear to have all remained intact and the crystal is still well bonded to the substrate. This suggests that the device degradation is confined to the first layers of the organic material at the interface with the dielectric.

To determine the cause of device degradation it is revealing to compare the behavior of single-crystal FETs fabricated using type I and type II Ta$_2$O$_5$. Fig. 3 shows the results of $I_D$-$V_G$ sweeps for two rubrene single-crystal FETs fabricated using the two different oxides. Similar to what we have shown for the tetracene FETs in Fig. 2, the FET on type I oxide exhibits a non-linear, non-monotonic $I_D$-$V_G$ relation up to the dielectric breakdown voltage (at $V_G \approx -80$ V for this sample). Conversely, for the rubrene FET on type II Ta$_2$O$_5$, $I_D$ scales linearly with $V_G$ in a large range of values above the threshold voltage, up to the maximum voltage reached in the experiment ($V_G = -100$ V, corresponding to a charge density of $\sim 5 \times 10^{13}$ holes/cm$^2$). For this FET, multiple mea-

FIG. 2: (a) Transfer characteristics of a FET on Ta$_2$O$_5$ ($V_D = -10$ V). The source and drain current ($I_S$ and $I_D$), are equal and opposite, since the leakage current $I_{\text{leak}}$ is orders of magnitude smaller than $I_D$ (see inset). Device degradation is responsible for the non-monotonic $I_D$-$V_G$ curve, resulting in the full suppression of $I_D$ at high $V_G$. (b) Transfer characteristics of a tetracene FET on ZrO$_2$ ($V_D = -10$ V). The $I_D$-$V_G$ relation is non-linear and $I_D$ and $I_S$ are lower in the second $V_G$ sweep (open circles) than in the first sweep (closed circles). $I_{\text{leak}}$ (triangles) is much smaller than $I_D$ and $I_S$. Note that the shape of the $I_D$-$V_G$ curves is characteristic for ZrO$_2$ and differs from that of Ta$_2$O$_5$. For comparison, (c) shows the transfer characteristics of a tetracene FET on SiO$_2$ for which $I_S$ and $I_D$ are linearly related to $V_G$ ($V_D = -10$ V).
measurements of the $I_D-V_G$ curve reproducibly give the same result. Note also, in the inset of Fig. 3 that the transfer characteristics of the rubrene FET on type II Ta$_2$O$_5$ are fully hysteresis-free, as is also typical for high-quality transistors fabricated on SiO$_2$. In short, contrary to what happens to devices based on type I Ta$_2$O$_5$, for FETs fabricated on type II Ta$_2$O$_5$ device degradation does not occur. Since the main difference between type I and type II Ta$_2$O$_5$ layers is the much higher level of leakage current observed in the type I layers, this observation suggests that the current leaking through the gate insulator is the cause for the device degradation.

To further investigate the origin of the FET degradation, we have also studied FETs fabricated on bilayers consisting of a 350 nm thick layer of type I Ta$_2$O$_5$ (ZrO$_2$) covered with a 25 nm thin top layer of ZrO$_2$ (type I Ta$_2$O$_5$), so that the organic crystal is in contact with the thin top layer. For these FETs, the shape of the $I_D$-$V_G$ curve is similar to that observed in FETs where the thin top layer is not present. These experiments indicate that the details of the device degradation are determined by the thick oxide layer and not by the material directly in contact with the organic crystals. This observation rules out the possibility that a chemical reaction between molecules and dielectric material is causing the device degradation and confirms the role of the leakage current, since in these oxide bi-layers it is the thick layer that determines the magnitude of $I_{\text{leak}}$.

We conclude that damage to the organic crystal induced by current leaking through the gate insulator is the cause for the device degradation. This conclusion is further supported by the absence of degradation in single-crystal FETs fabricated on SiO$_2$, in which the leakage current is undetectably small. It is also consistent with the larger degradation observed in type I Ta$_2$O$_5$ FETs as compared to ZrO$_2$ devices, since the leakage current through ZrO$_2$ is typically almost an order of magnitude less than in type I Ta$_2$O$_5$.

It is worth noting that degradation occurs even when the leakage current is several orders of magnitude lower than the source-drain current. Specifically, our data quantitatively show that in organic single-crystal FETs gate leakage currents larger than approximately $10^{-9}$ A/cm$^2$ systematically result in irreversible device degradation. This conclusion poses a clear constraint on the design of properly functioning single-crystal FETs. It is possibly also relevant for organic thin-film transistors, as it suggests that the gate leakage current is an important factor in determining the device lifetime.

In conclusion, we have shown that leakage current from the gate electrode causes irreversible degradation of organic single-crystal FETs, even when it is orders of magnitude smaller than the source-drain current. This poses a clear constraint for the design of single-crystal transistors currently used to investigate the intrinsic electronic properties of organic semiconductors. As a byproduct of this work, we have successfully fabricated single-crystal devices operating up to a charge density of at least $5 \times 10^{13}$ carriers/cm$^2$ ($\sim 1$ carrier per 10 molecules), which will enable the investigation of organic single-crystal FETs at high carrier density.

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The microscopic origin of the difference in leakage current between Type I and Type II Ta$_2$O$_5$ is probably due to the larger deposition rate that can be achieved in sputtering from a metal target, which results in the inclusion of less impurities in the sputtered layers.

The specific microscopic process responsible for the degradation of the organic material remains to be understood. One possible mechanism is that high-energy electrons leaking through the gate insulator physically break individual molecules at the crystal surface, thus causing the appearance of a very large number of traps in the FET active regions.

A more precise quantification is difficult because an unknown fraction of leakage current flows directly to the source or drain contacts and does not cause damage to the organic crystal.