Reproducible Low Contact Resistance in Rubrene Single-Crystal Field-Effect Transistors with Nickel Source and Drain Electrodes

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Abstract

We have investigated the contact resistance of rubrene single-crystal field-effect transistors (FETs) with Nickel electrodes by performing scaling experiments on devices with channel length ranging from 200 nm up to 300 µm. We find that the contact resistance can be as low as 100 Ωcm with narrowly spread fluctuations. For comparison, we have also performed scaling experiments on similar Gold-contacted devices, and found that the reproducibility of FETs with Nickel electrodes is largely superior. These results indicate that Nickel is a very promising electrode material for the reproducible fabrication of low resistance contacts in organic FETs.
The possibility to downscale organic field-effect transistors (FETs) is currently hindered by the high contact resistance present at the interface between the metal electrodes and the organic semiconductor \[1\]. One of the main experimental problems in the study and optimization of the contact resistance originates from the observed irreproducibility. In spite of the large effort put in the investigation of contact effects \[1, 2, 3, 4, 5, 6\], the reason for both the high values and the irreproducibility of the contact resistance are not currently understood. Many different phenomena are likely to play an important role, including the presence of grain boundaries at the metal/organic interfaces, the interface fabrication process (e.g., metal diffusion into the organic semiconductors and extrinsic damage introduced during the device assembly process), fluctuations in the work function of the metal electrodes, etc. Currently, the problem seems to be particularly severe for oligomer-based devices. Whereas for FETs based on a number of different polymers it has been found that the contact resistance scales linearly with the carrier mobility \[7\], for transistors based on oligomers a very broad range of contact resistance values have been measured on identically prepared devices, and no systematic behavior has been observed \[6\].

To address the issue of contact resistance in oligomer transistors, we have recently started the investigation of organic single-crystal FETs with different metal contacts. Single-crystal devices are particularly advantageous for this purpose because their electrical characteristics exhibit an excellent level of reproducibility from sample to sample \[8\]. This is crucial for a reliable comparison of FETs with different channel length, i.e. to perform scaling experiments from which the value of the contact resistance can be extracted.

In this paper we focus on rubrene single-crystal FETs with Nickel electrodes. Nickel was chosen because, although it oxidizes in air, its native oxide is conductive and has a work-function of 5.0 eV \[9\], ideally suited to inject carriers into the highest occupied molecular orbital of many molecular semiconductors. By performing a conventional scaling analysis \[6, 7, 10\] of the electrical characteristics of these devices we extract the value of the contact resistance. We find values of \(R_C\) as low as 100 Ωcm, i.e. 50 times smaller than in the best oligomer FET reported to date \[6\]. The spread in values in the contact resistance measured on transistors fabricated on the same crystal is small (less than a factor of 2); devices fabricated on different crystals exhibit a somewhat large spread, ranging from 100 Ωcm to 1.5 kΩcm (and typically between 200 Ωcm and 1 kΩcm), but still considerably smaller than what has been observed so far in oligomer FETs. For comparison, we have also investigated
a number of single-crystal FETs contacted with gold electrodes, the material commonly used for the fabrication of contacts in organic transistors, and found a considerably lower reproducibility level. This indicates that Nickel is a very promising material for the fabrication of contacts for organic transistors, even though the surface of the electrodes oxidizes. We note that Nickel is also advantageous as compared to gold because it is more mechanically robust, which should minimize the possibility of (electro)migration into organic materials during device operation, and cheaper.

The FET fabrication is based on electrostatic bonding of rubrene single crystals to a doped silicon substrate (acting as a gate) covered with a 200 nm- thick thermally grown SiO$_2$, with prefabricated source and drain contacts (see Ref.\[11\] for details). The contacts are prepared by conventional optical or e-beam lithography, nickel electron-beam evaporation (20 nm), and lift-off. The rubrene crystals are separately grown by means of a vapor phase transport technique\[12\]; they are millimeters long and their width and thickness are respectively of the order of 100 $\mu$m and 1 $\mu$m. The device layout (see Fig.\[1\]) is such that FETs with different channel lengths are fabricated on the same single crystal. Many different samples were studied with channel length ranging from 200 nm to 300 $\mu$m. Prior to the crystal adhesion, an oxygen plasma treatment is performed to remove residues of resists possibly still present on the SiO$_2$ surface. Although the exposure of the electrodes to oxygen plasma contributes to the oxidation of the Nickel surface, it does not preclude the realization of reproducible, low-resistance electrodes.

In the linear regime of transistor operation the total device resistance $R_T(L)$ can be written as \[6, 7, 10\]:

$$R_T(L) = R_{ch}(L) + R_C,$$

Here

$$R_{ch} = \frac{L}{W C_i (V_G - V_{TH})} \frac{1}{\mu}$$

is the channel resistance and $R_C$ a length independent contact resistance, $C_i$ is the capacitance of the insulating layer per unit area, $V_G$ and $V_{TH}$ are the gate and the threshold voltage, $W$ is the channel width, and $\mu$ the hole mobility. The contact resistance is obtained by extrapolating the experimental data to zero channel length. The slope of the $R_T$-vs-$L$ curves also permits to extract the carrier mobility. The comparison of the mobility value obtained from this slope with the one obtained from the usual formula for the linear regime
of the individual FETs

\[ \mu = \frac{L}{W C_i V_{DS} \partial I_{DS} \partial V_G} \]  

(3)

is used as a consistency check of our analysis.

Fig. 1 shows the electrical characteristics measured on one of the FETs that we have investigated and it is typical for all our Ni contacted devices. The data have been measured with the FETs in high vacuum \((p < 10^{-6} \text{ mbar})\) and dark, using an Agilent E5270A or a HP 4192A parameter analyzer. Usually, no hysteresis is observed in the \(I_{DS} - V_{DS}\) plot at fixed gate voltage and in the \(I_{DS} - V_G\) at fixed source drain bias. The linearity of the \(I_{DS} - V_{DS}\) at low bias gives a first indication of a good contact quality.

The scaling of the total device resistance \(R_T\) versus device length \(L\) is shown in Fig. 2 for different values of the gate voltage \(V_G\), with \(V_{DS} = -1 \text{ V}\), for a sample with channel length in the range 20 -200 \(\mu\)m. Clearly, \(R_T\) does scale linearly with \(L\), implying that for a given device the contact resistance \(R_C\) is approximately the same irrespective of the channel length. The value of \(R_C\) is then given by the intercept at \(L=0\). To compare the behavior of devices fabricated on different crystals we normalize the contact resistance to the channel width, i.e. we consider \(R_C^* = R_C W\) [1, 6]. For all different samples (in total, approximately 50 individual FETs were measured) we find values of \(R_C^*\) in between 100 \(\Omega\text{cm}\) and 1.5 k\(\Omega\text{cm}\), and most typically in between 200 \(\Omega\text{cm}\) and 1 k\(\Omega\text{cm}\), at \(V_G = -30 \text{ V}\), usually only very weakly dependent on gate voltage.

We have also analyzed the spread in contact resistance values for FETs fabricated on the same crystal, by looking at devices with \(L\) ranging from 200 nm to a few microns. Because the rubrene crystals have a high mobility \((2-6 \text{ cm}^2/\text{Vs})\), the contact resistance exceeds the channel resistance in devices whose channel length is less than approximately 5-10 \(\mu\)m. For these devices, the total resistance is essentially independent of channel length, as shown in Fig. 2c. These data also show that for FETs fabricated on the same crystal, the spread in contact resistance values is less than a factor of two. Thus, both for short and long channel devices, we conclude that the values of \(R_C^*\) in Nickel-contacted Rubrene single crystal FETs are up to 50 times smaller than the smallest contact resistance (5 k\(\Omega\text{cm}\)) reported to date for oligomer-based FETs [6], and that they exhibit a drastic improvement in reproducibility as compared devices studied in the past.

For devices with a channel length of 100 \(\mu\)m or longer, we have calculated the value of mobility from the FET characteristics using Eq. 3, as well as from the scaling analysis using
Eq. 2. The comparison of the values obtained in these two different ways (see inset of Fig. 3) exhibits a remarkable agreement. This agreement indicates the consistency of our analysis and gives full confidence on the quantitative values obtained for the contact resistance.

To understand if the low values and the reproducibility of the contact resistance are due to the use of Nickel electrodes, or if they are just a consequence of using high-quality organic single crystals for the device fabrication, we have performed a scaling analysis also for several gold-contacted single crystal FETs. In all of these gold-contacted devices the mobility obtained via Eq. 3 (for long channel devices, $L > 100 \, \mu m$) ranged from 2 to 6 $\text{cm}^2/\text{Vs}$, depending on the crystal [13]. This indicates that the crystal quality is the same for gold and Nickel-contacted FETs. However, we found that in FETs with gold electrodes the fluctuations in contact resistance are much larger and in most cases prevent the observation of a clear scaling between $R_T$ and $L$, for channel lengths comparable to or smaller than 50-100 $\mu m$. This is illustrated in Fig 2b that shows the data for the gold-contacted devices which, among all devices measured, exhibited the best $R_T(L)$ scaling: as it is clear the fluctuations in measured resistance are much larger than for the Nickel-contacted devices (Fig 2b and c). As a consequence of poor scaling, the data on gold-contacted transistors do not allow a precise determination of $R_C$ but only a rough estimate of the lower limit, 5 $\text{k}\Omega\text{cm}$, with a spread of several times this value (even for FETs fabricated on the same rubrene crystal).

From the above comparison, we conclude that Nickel does perform better than gold as electrode material and that the reproducibility in the contact resistance is not only due to the use of single-crystalline material for the FET fabrication. As gold has been used for contact fabrication in most of the organic FETs fabricated in the past also in virtue of its chemical inertness, the fact that Nickel does oxidize in air makes our findings particularly unexpected. Nevertheless, the low contact resistance values can be explained in terms of the work function of oxidized Nickel that has been measured to be equal to 5.0 eV[6] -ideal for hole injection into organic semiconductors- and by the fact that non-stechiometric NiO$_x$ is a reasonably good conductor. In this regard, it is also worth noting that recently NiO$_x$ contacts have shown promising results as hole injectors in organic light emitting diodes [14]. Why oxidized Nickel performs better than gold [15], which is a better conductor and has a comparable work function value, is less clear: for its technological relevance, this issue deserves additional investigations.
In conclusion, we have performed a scaling analysis of the electrical characteristics of rubrene single-crystal FETs to show that nickel can be used to fabricate source and drain electrodes with an unprecedented low contact resistance and excellent reproducibility.

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[15] Note that very recently pentacene thin-film FETs with evaporated NiOx contacts have been claimed to have better performance than identically prepared devices with gold contacts. See J. Lee, D. K. Hwang, J. M. Choi, K. Lee, J. H. Kim, S. Im, J. H. Park and E. Kim, Applied Physics Letters 87, 023504 (2005). In this work, however, the value of the contact resistance was not measured.
FIG. 1: Typical transistor characteristics measured on a rubrene single-crystal FET with Ni source-drain electrodes. The inset shows a top view of one of the devices used in our investigation (for this device the crystal width $W$ is $35 \mu m$).
FIG. 2: (a) Scaling of the device resistance for Nickel contacted devices as a function of channel length for different values of the gate voltages (W = 35 μm). The intercept at L = 0 gives the contact resistance. (b) Similar scaling curve for a gold-contacted FET: it is visible that the deviations from linear scaling are larger in this devices as compared to Nickel-contacted devices. In other gold-contacted FETs, the magnitude of the fluctuations was larger than for the sample whose data are shown here. (c) Normalized resistance measured on a Ni-contacted FETs fabricated on the same rubrene crystal. $R_T$ does not depend on $L$ because for $L < 2\mu m$ the channel resistance is negligible with respect to the contact resistance. In all panels, the lines are a guide to the eye.
FIG. 3: Gate voltage dependence of the normalized contact resistance $R_C^*$ for four of the samples studied. The insert shows the gate voltage dependence of the mobility $\mu$ determined from Eq. 3 (full circles) and from scaling $R_T(L)$ using Eq. 2 (open diamonds). The vertical line denotes the beginning of the linear regime. For all the measurements $V_{DS} = -1$ V.