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Bias-dependent contact resistance in rubrene single-crystal field-effect transistors

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The authors report a systematic study of the bias-dependent contact resistance in rubrene single-crystal field-effect transistors with Ni, Co, Cu, Au, and Pt electrodes. They show that the reproducibility in the values of contact resistance strongly depends on the metal, ranging from a factor of 2 for Ni to more than three orders of magnitude for Au. Surprisingly, field-effect transistors with Ni, Co, and Cu contacts exhibit an unexpected reproducibility of the bias-dependent differential conductance of the contacts once this has been normalized to the value measured at zero bias. This reproducibility may enable the study of microscopic carrier injection processes into organic semiconductors. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2741411]

Considerable improvements in the material control of organic thin films are now enabling the reproducible, low-cost fabrication of organic field-effect transistors (FETs) with mobility values in the range of $0.1-1\ \text{cm}^2/\text{V s}$.[1] These values are sufficient for the development of applications in the field of plastic electronics.[2] However, the fabrication of high-quality electrical contacts for organic transistors has not progressed comparably,[3] and contacts are now posing limits to the performance of organic FETs. Specifically, with mobility values in between 0.1 and $1\ \text{cm}^2/\text{V s}$, the contact resistance—typically larger than $1\ \text{k}\Omega\ \text{cm}$ even in the best devices—limits the transistor performance as soon as the channel length becomes smaller than $\simeq 10\ \mu\text{m}$,[4] preventing the possibility of device downscaling. Irreproducibility makes the situation even worse: for gold-contacted pentacene thin-film FETs, for instance, the spread in contact resistance values was recently observed to exceed three orders of magnitude (from $2\ \text{k}\Omega\ \text{cm}$ to more than $1\ \text{M}\Omega\ \text{cm}$).[5] The current lack of understanding of the microscopic carrier injection[6] processes from a metal electrode into an organic semiconductor does not help us in determining the causes of the observed irreproducibility and more systematic experiments are needed.

Here we report systematic transport measurements of rubrene ($C_{42}H_{28}$) single-crystal FETs with electrodes made of five different metals (Ni, Co, Cu, Au, and Pt). All the transistors have been fabricated with a sufficiently short channel length, so that the total device resistance is entirely dominated by the contacts. By studying more than 250 contact-dominated devices, we have collected enough statistics to determine the average contact resistance, its spread in values, as well as its bias dependence. We find significant differences between the different metals. In particular, for nickel—which exhibits the lowest resistance—the spread is only a factor of 2, for cobalt and copper slightly more than one order of magnitude, and for gold more than three orders of magnitude (platinum seems to behave similarly to gold, but the number of devices tested was not sufficient to make more quantitative statements). We also find that for Ni, Cu, and Co (but nor for Au and Pt) the bias dependence of the contact resistance normalized to the resistance measured at zero bias exhibits an excellent reproducibility and can be interpreted in terms of two back-to-back Schottky diodes connected in series. Our results suggest that organic single-crystal FETs with Ni, Co, and Cu contacts may be suitable for the investigation of the microscopic carrier injection processes at a metal/organic interface.

Some of the metals (i.e., Cu and Co) investigated here have not been used previously for the fabrication of organic FETs. In fact, the vast majority of past experiments have relied on noble metal electrodes, especially gold, whose choice is motivated by the high value of their work function and by the stability against oxidation in air. These criteria, although plausible, have never been thoroughly investigated. Our recent and unexpected finding of the record-low contact resistance in devices with nickel electrodes ($100\ \Omega\ \text{cm}$)[7] clearly underscores the importance to explore a broader class of materials.

The rubrene single crystal FETs are fabricated by lamination of thin ($\approx 1\ \mu\text{m}$ thick), free standing crystals grown by vapor phase transport, onto a highly doped Si substrate, covered by a 200 nm thick thermally grown SiO_2 layer with predefined metal electrodes (see Ref. 8). The contacts are fabricated by means of electron-beam lithography, evaporation, and lift-off, and their geometry is chosen so that many FETs with channel length varying from 200 nm to 50 μm can be fabricated on the same crystal (see inset in Fig. 1). Our earlier studies have shown that the room-temperature carrier mobility in rubrene single-crystal FETs with SiO_2 gate dielectric is narrowly spread around $4\ \text{cm}^2/\text{V s}$,[9] which allows us to estimate the maximum channel length L for which the channel resistance can be neglected with respect to the contact resistance. Specifically, for FETs with nickel contacts (whose resistance is normally lower than $1\ \text{k}\Omega\ \text{cm}$) we have confined our measurements to devices with L smaller than 2 μm. For devices with electrodes made of the other materials, where the contact resistance is higher, devices with channel lengths up to $\simeq 20\ \mu\text{m}$ have also been used. In all cases, therefore, the measured device resistance corresponds to the sum of the source and drain resistances.

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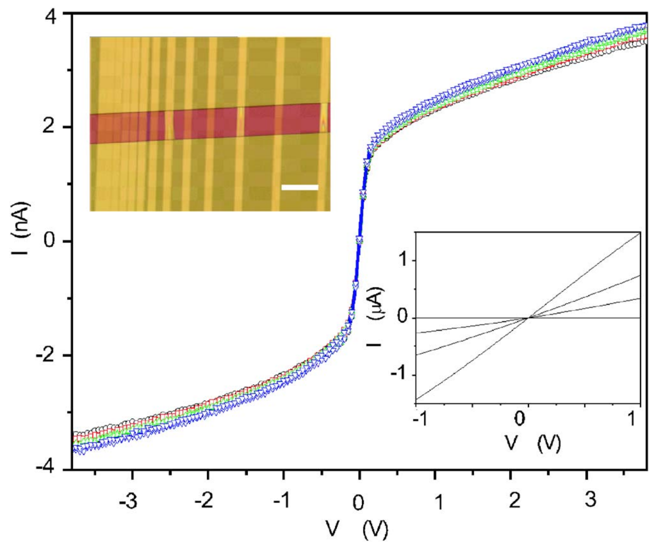

FIG. 1. (Color online) *I-V* curves for a contact-dominated FET (L=5 μm for four different gate voltages, V_G=−20, −30, −40, and −50 V) showing nonlinear behavior, different from the usual FET characteristic reported in the inset (shown in the lower inset from measurements on long channel device). Top left inset: Optical microscope image of one of our devices (the white bar is 50 μm long).

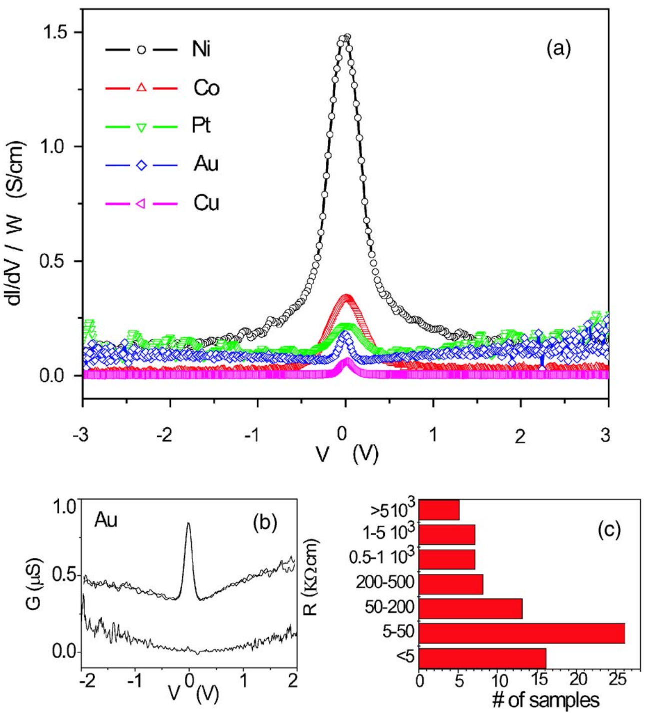

FIG. 2. (Color online) (a) Differential conductance normalized to the crystal width (W) measured on FETs with different metal electrodes. (b) Differential conductance for two different short-channel gold-contacted FETs. (c) Histogram showing the spread in contact resistance for Au electrodes.

Figure 1 shows the *I-V* characteristics of a Cu-contacted FET with L=5 μm, measured[10] at different values of the gate voltage (much larger than the threshold voltage). The curves are essentially independent of the gate voltage and exhibit a very pronounced non-linear increase of the source-drain current at low bias. These *I-V* characteristics are markedly different from those of conventional transistors where the resistance is dominated by the channel, as illustrated in the inset of Fig. 1. A nonlinear and gate-voltage independent *I-V* curve similar to that shown in Fig. 1 has been observed in short-channel devices fabricated with all the different metal electrodes, and to analyze this behavior in more detail we look at the differential conductance dI/dV of the devices [obtained by numerical differentiation, see Fig. 2(a)]. In the differential conductance, the nonlinearity present in the *I-V* characteristics produces a narrow peak around zero bias on a voltage scale comparable to kT/e, the precise value being different for the different metals.

In order to compare the contact properties of FETs contacted with the five different metals we have carefully analyzed the reproducibility of the measured differential resistance. We find that the level of reproducibility depends on the specific metal used. The differential conductance measured on two different short-channel gold-contacted FETs is shown in Fig. 2(b), which illustrates the poor reproducibility of these devices. In fact, for Au contacts (and similarly for Pt) the bias dependence of the differential conductance exhibits large differences in different samples and a zero-bias peak is observed only in a few devices. At the same time, the absolute value of the resistance measured at low bias is spread over three orders of magnitude [see Fig. 2(c)].

The situation is very different for Ni-, Co-, and Cu-contacted transistors. Figures 3 and 4 show the differential conductance curves for many FETs with nickel and copper electrodes, normalized to the zero-bias value. Remarkably, all curves fall nearly on top of each other, indicating a good reproducibility of the bias dependence of the contact resistance. The histograms shown in the insets of Figs. 3 and 4 quantify the reproducibility in the absolute (zero-bias) value of the contact resistance. For nickel the resistance of the majority of devices is $dV/dI(V=0)=500\pm250$ Ω cm; for the few devices for which $dV/dI(V=0)>1000$ Ω cm the larger resistance is likely to originate from an imperfect lift-off process during the electrode fabrication, causing a poor adhesion of the crystal to the metal surface. For copper—and similarly for cobalt (data not shown)—the spread in contact resistance is between one and two orders of magnitude (still considerably smaller than for gold), which makes the reproducibility of the bias dependence even more surprising. Overall, it is a remarkable finding that materials such as Ni,

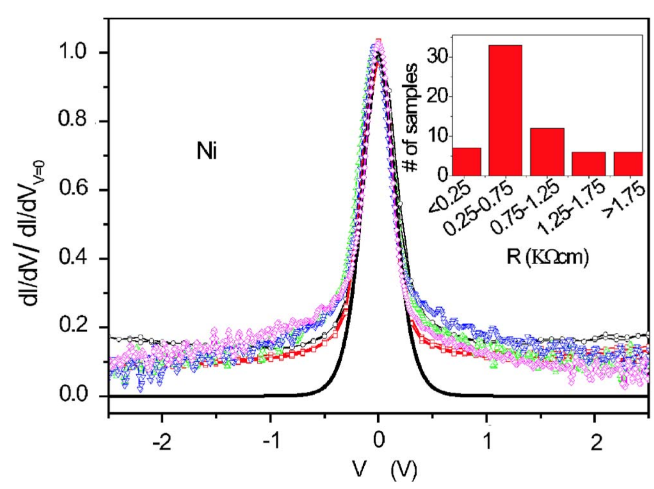

FIG. 3. (Color online) Normalized differential conductance for eight different FETs with Ni electrodes (open symbols) exhibiting reproducible behavior. The continuous line is a plot of the differential conductance of two Schottky diodes in series obtained from Eq. (2). Inset: Histogram showing the spread in contact resistance for Ni electrodes.

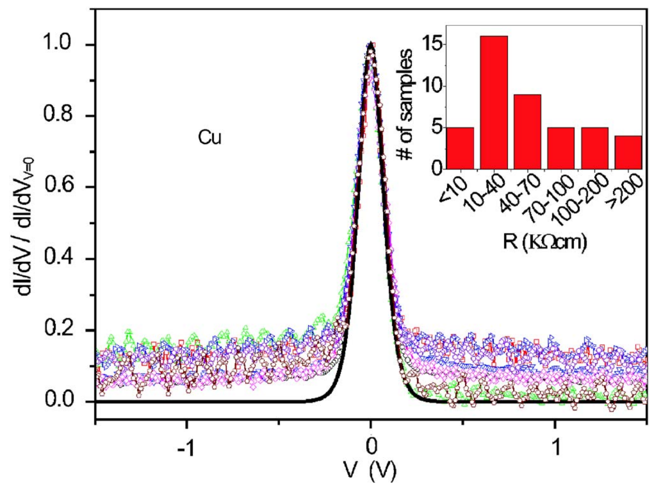

FIG. 4. (Color online) Normalized differential conductance for ten different FETs with Cu electrodes (open symbols) exhibiting reproducible behavior. The continuous line is a plot of the differential conductance of two Schottky diodes in series obtained from Eq. (2). Inset: Histogram showing the spread in contact resistance for Cu electrodes.

Co, and Cu, whose surfaces oxidize during fabrication, lead to an enhanced reproducibility. The reason for this is currently unclear. We point out, however, that the difference between these materials and gold is not likely to be the value of their work function—normally invoked as the main parameter determining the quality of the contacts—since the work functions of Au and oxidized Ni are very similar (5.1 and 5.0 eV,[11] respectively).

As an initial step in interpreting the experimental data, we model our devices as two oppositely biased Schottky diodes connected in series (corresponding to the metal/organic and organic/metal interfaces at the source and drain contacts). The simplest expression for the current through a diode[12] reads:

$$I(V) = I_0(e^{eV/nkT} - 1), \tag{1}$$

where n is the so-called ideality factor and I_0 is taken to be constant (i.e., independent of V). The resulting I-V curve for two back-to-back diode I-V curves then is

$$I(V) = I_0 \tanh\left(\frac{eV}{2nkT}\right). \tag{2}$$

The continuous lines in Figs. 3 and 4 are plots of the differential conductance calculated by differentiating this equation, from which we see a qualitative agreement between the data and the back-to-back Schottky diode picture. The reproducibility of the data is sufficient to discriminate quantitatively between the behavior of nickel, for which the width of the peak corresponds to an ideality factor of $n \simeq 3$, and for copper, where $n \simeq 1$. The deviation at high bias, i.e., the fact that the measured differential conductance is higher than what is expected, originates from having assumed that I_0 is a constant, whereas in reality I_0 depends on bias. Within conventional models of metal/semiconductor interfaces the bias dependence of I_0 may originate from different microscopic phenomena, such as a bias-induced lowering of the Schottky barrier (i.e., the Schottky effect), diffusion limited transport, tunneling, etc..[12] To discriminate between these mechanisms and to determine whether one of them can explain the observed I-V characteristics of the contacts, measurements at different temperatures are needed. Work is ongoing in this direction, and preliminary results seem to indicate that for Cu contacts the inclusion of the Schottky effect should lead to quantitative agreement with data, whereas for nickel tunneling may also be important. We also hope that the analysis of these results will provide indications as to the microscopic origin of the irreproducibility in the contact resistance of nominally identical samples.

In conclusion, we have shown that Ni, Cu, and Co electrodes enable a superior reproducibility of the contact characteristics as compared to metals used in the past and open the possibility for a controlled study of microscopic processes of carrier injection into organic semiconductors.

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