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# Charge carrier injection and ambipolar transport in C<sub>60</sub>/CuPc organic semiconductor blends

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**Abstract.** Ambipolar organic field-effect transistors with a mixed active layer of n-conducting fullerene and p-conducting copper-phthalocyanine were prepared. The influence of the mixing ratio and the preparation conditions on the mobilities and the threshold voltages was determined for both charge carrier types. Hole accumulation in the phthalocyanine at the fullerene/phthalocyanine interface is observed. A strong correlation between contact resistance and mobility indicates that carrier injection is diffusion limited.

## 1. Introduction

Mixed films of organic materials are often used as active layer in photovoltaic cells, as host-guest systems in organic light emitting diodes and for conductivity doping of organic materials. Another recent application is the usage in ambipolar organic field-effect transistors (OFET), e.g. as light emitting OFETs [1] and as inverters in electronic circuits [2, 3].

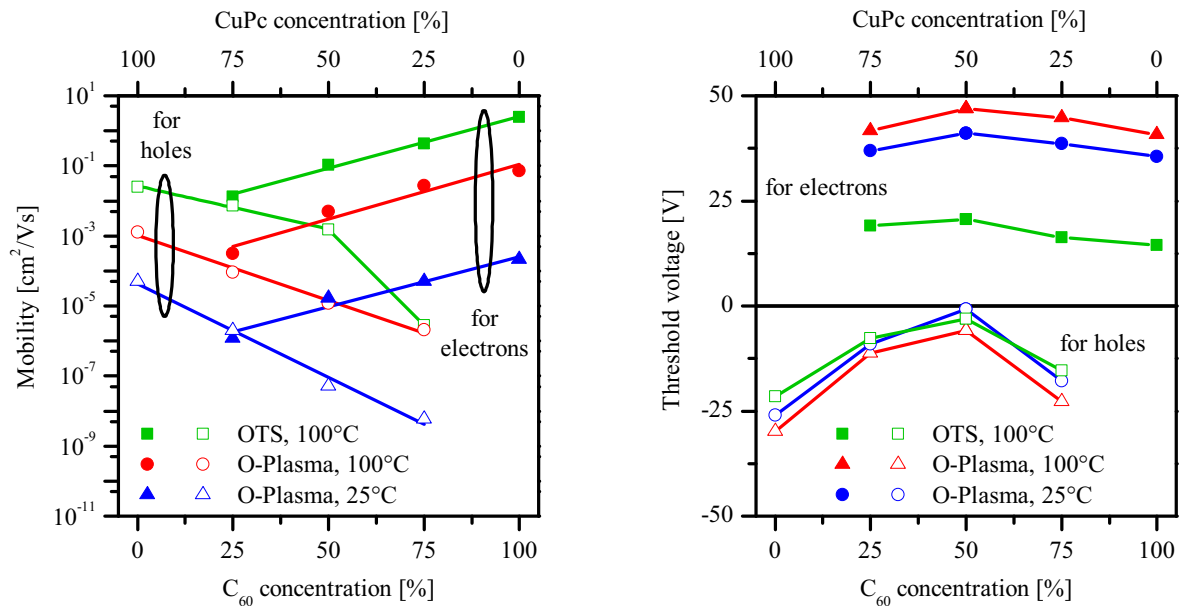
In this contribution the influence of the interfaces on the transport properties will be investigated in dependence on the mixing ratio of blends from n-conducting fullerene and p-conducting copper-phthalocyanine. The transport properties are affected by both the insulator/semiconductor interface (here the surface treatment of the Si/SiO<sub>2</sub> substrate) and the substrate temperature during evaporation of the organic blend. The injection is determined by the metal/semiconductor interface and was analysed by measuring the injection barrier and the contact resistance.

## 2. Mobility and threshold voltage

The preparation of the devices by co-evaporation on pre-structured thin-film transistor substrates (silicon oxide as gate insulator and gold as electrodes in bottom contact and bottom gate geometry) and the analysis in the saturation regime are described elsewhere [3]. Additionally a treatment of the silicon oxide surface by octadecyltrichlorosilane (OTS) is used [4].

Figure 1 shows the mobility and the threshold voltage determined from the saturation regime as a function of concentration in the blend for different preparation conditions. The substrate temperature during evaporation of the blends was 25°C and 100°C for the O-plasma treated silicon oxide surface. The surface chemistry was modified additionally for the 100°C substrate temperature by silanisation of SiO<sub>2</sub> with OTS.

An exponential decrease of both electron and hole mobility is observed upon dilution of the corresponding conducting material with the other species. An increased mobility is found for the



**Figure 1.** Mobility (left) and threshold voltage (right) determined from saturation regime as function of concentration, substrate temperature and substrate treatment. The filled symbols are related to the electron transport, the open symbols to the hole transport.

higher substrate temperature and a further increase is realised by lowering the surface energy with OTS. This increase of mobility is reported for unipolar OFETs [5] and is also valid for these blends. For all treatments balanced mobilities are found at about 25%  $C_{60}$  content. This balance is necessary e.g. to realize symmetric ambipolar inverters [3].

The threshold voltage shows a dependence on the mixing ratio for the hole channel, but not for the electron channel. By contrast, the threshold voltage for the electron channel changes with the preparation conditions, especially with the OTS passivation of the silicon oxide. The reason is that the O-plasma treated oxide surface contains OH-groups which are acting as electron traps and increase the threshold voltage for electrons [6]. By surface treatment with OTS the traps are passivated and the threshold voltage decreases. The change of threshold voltage for the hole transport with the concentration is related to the organic/organic interface in the  $CuPc/C_{60}$  blend [3] and can be explained by a hole accumulation at the  $CuPc$  side of the  $C_{60}/CuPc$  interface [7]. Because there is no charge transfer from  $CuPc$  to  $C_{60}$  in the ground state this shift should be related to a displacement of charges within the  $CuPc$  molecules in the presence of  $C_{60}$  as demonstrated by calculations [8]. This charge displacement is independent on the preparation conditions. The threshold voltage shift is related to the interface charge [9] at the organic/organic interface in the proximity of the insulator by:

$$\Delta V_T = \frac{e \cdot N_{If}}{C_{Ox}}. \quad (1)$$

By modeling the molecular packing [3] a charge accumulation of about 0.012 charges per molecule is estimated. The calculated charge transfer in an ideal complex of one  $C_{60}$  and one  $CuPc$  molecule is 0.06 charge per molecule [8], 5 times higher than determined here.

### 3. Contact resistance

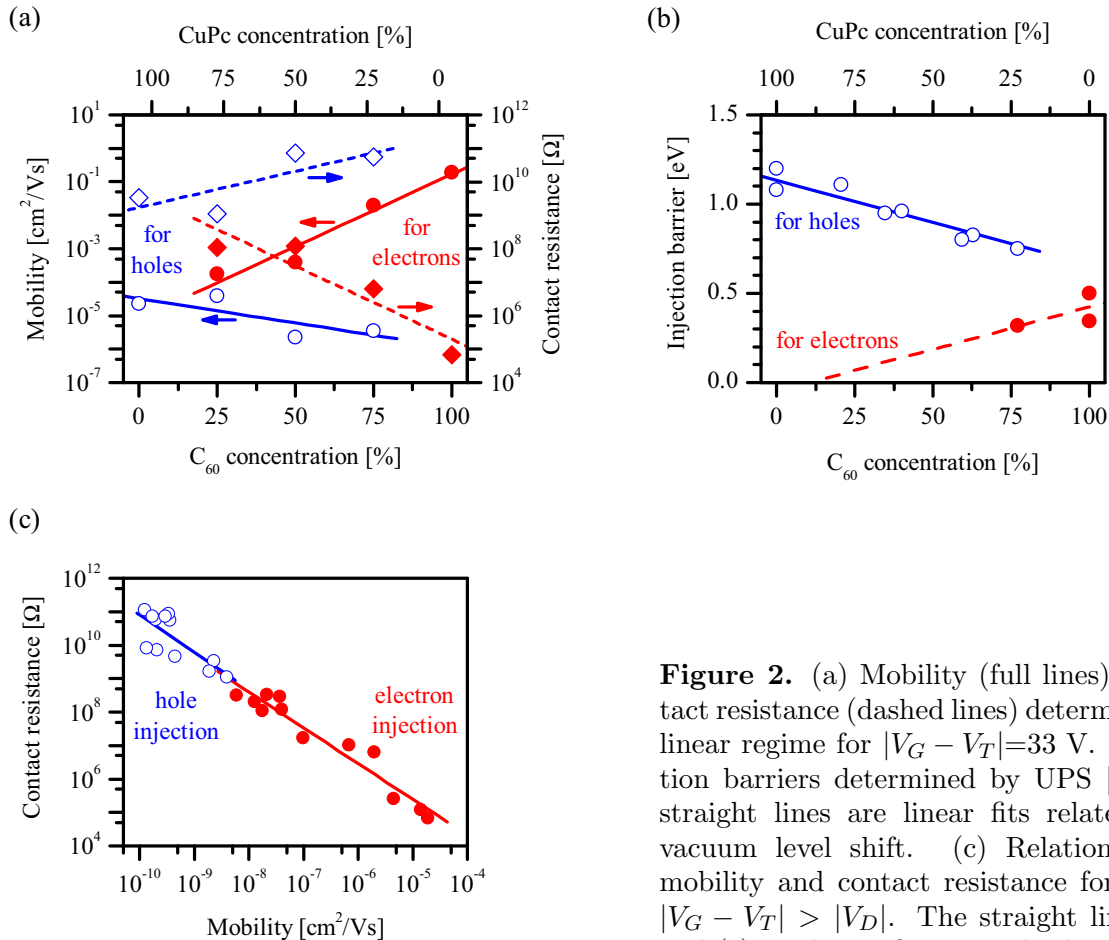
In contrast to the first part of this paper, we now use transfer curves in the linear range for analysing the interdependence of mobility and contact resistance [11]. Therefore the drain voltage in the Shockley equation is replaced by the drain voltage corrected by the contact resistance ( $V_D \rightarrow V_D - I_D \cdot R_C$ ). As result the drain current is given by

$$I_D = \frac{(W/L)C_i\mu(V_G - V_T)V_D}{1 + (W/L)C_i\mu R_C(V_G - V_T)}. \quad (2)$$

Using the channel conductance  $g_d = I_D/V_D$  and the transconductance  $g_m = \partial I_D/\partial V_G$  the mobility  $\mu$  and the contact resistance  $R_C$  were calculated from

$$\sqrt{\mu}(V_G - V_T) = \frac{g_d}{\sqrt{g_m}} \cdot \sqrt{\frac{LV_D}{ZC_i}} \quad \text{and} \quad R_C = \frac{1}{g_d} - \frac{L}{W\mu C_i(V_G - V_T)}. \quad (3)$$

In this calculation the dependence of  $\mu$  and  $R_C$  on  $V_G$  is neglected for the derivatives. To minimize the error of this simplification the mobilities and the contact resistances are plotted in fig. 2a for  $V_D = 10$  V at a constant  $|V_G - V_T| = 33$  V (using O-plasma treatment and 100°C substrate temperature). Once again, one observes decreasing mobilities with decreasing concentration of the transport material like in the saturation regime shown in fig. 1a. The contact resistance increases with diluting the respective transport material. This behaviour is



**Figure 2.** (a) Mobility (full lines) and contact resistance (dashed lines) determined from linear regime for  $|V_G - V_T| = 33$  V. (b) Injection barriers determined by UPS [10]. The straight lines are linear fits related to the vacuum level shift. (c) Relation between mobility and contact resistance for different  $|V_G - V_T| > |V_D|$ . The straight lines in (a) and (c) are linear fits to guide the eyes.

in contrast to the injection barrier shown in fig. 2b (as determined from the difference between the Fermi level and the bulk transport edge by ultraviolet photoelectron spectroscopy) which decreases with increasing amount of the respective transport material. For this analysis interface effects (band bending and interface dipole) were neglected.

Figure 2c shows the contact resistance vs. mobility for all presented mixing ratios and for different charge carrier densities (related to different values of  $|V_G - V_T|$ ). A reciprocal relation  $R_C \sim \mu^{-1}$  is observed indicating that in the presented system the mobility limits the injection of charge carriers. This behaviour can be explained by diffusion limited injection [9] following the equation

$$j_{inj} \sim \mu \cdot \exp \left[ -\frac{\Phi_B}{kT} \right]. \quad (4)$$

The same was observed in diodes [12] varying the mobility by mixing semiconducting and insulating molecules and in unipolar OFETs [13] varying the mobility by the charge carrier concentration.

#### 4. Summary

We have investigated charge carrier mobilities, threshold voltages and contact resistances in field-effect transistors with blends of C<sub>60</sub> and CuPc as active semiconducting layer. For all mixing ratios ambipolar transport was observed, however, the magnitude of electron and hole mobilities strongly depends on the mixing ratio and the preparation condition. demonstrating a crucial role of the organic/insulator interface. The observed threshold voltage shifts indicate that hole accumulation is present in CuPc in contact with C<sub>60</sub> at the organic/organic interface. In spite of decreasing injection barrier for mixed layers the contact resistance increases inversely proportional to the mobility. This shows that the injection of charge carriers into these blends is diffusion limited.

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