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Injection-limited current in a polymeric heterojunction

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Abstract:

This work describes the current over an interface between two different polymeric semiconductors. The interface barrier amounts to 0.7 to 1.0 eV, which is orders of magnitude larger than the thermal energy. It is demonstrated that the current across the interface is injection limited. Furthermore, it is observed that the electrical characteristics for such an organic organic interface are completely different from those for a metal organic interface.

1. Introduction

The organic light-emitting diode (OLED) area is rapidly growing. Several companies have already introduced their products on the market. The characteristic properties are low weight and cheap manufacturing. Moreover, OLEDs have a wide viewing angle and intense pure colors. In addition, polymer light emitting diodes (PLEDs) have a large flexibility, which make them ideal candidates for portable applications. The PLEDs have a large difference in hole and electron mobility, shifting the recombination towards the cathode interface. Losses occur due to exciton quenching to the metal cathode, and a substantial amount of efficiency is lost.¹ Already for a long time solutions have been suggested, and one of the most efficient is to confine charges in the semiconductor away from the interface.² This can be achieved by separating the transport of the charge carriers from the recombination. The most simple example of such a confinement is an organic organic heterojunction with a band-offset to prevent the charges from transport over the interface. In this way, the recombination zone is more or less confined to the small interface area itself.

The organic-organic interface (OOI) has the remarkable property of a defect free heterojunction.³ This makes it an ideal device to investigate the electrical properties, such as the charge transport and diffusion over such an interface, and the charge injection for interfaces with a band-offset.

The charge injection over an OOI will be different from charge injection over a metal – organic interface (MOI) for two main reasons:⁴

First of all, the charge reservoir in the injecting layer cannot be thought to be infinite, and will certainly influence the injection rate.

Secondly, the OOI barrier is not subject to image force lowering, as the charge density in the injecting layer is too low to induce an image charge, while also the dielectric relaxation time of an organic semiconductor is much too long.⁵ This has consequences for the electric field dependence of the charge injection. The back-flow of injected charge carriers over a MOI is governed by the electric field close to the metal that counteracts the injection of charges.⁶ This will reduce the charge injection at low electric fields due to a large back flow of carriers by the image force, which is described by the Onsager escape probability. However, for a OOI this back-flow of carriers cannot be due to the image force, and must be taken into account by the backward hopping probability. This results in a completely different field dependence.⁴

To investigate the charge injection over an OOI, a bi-ethyl hexyl poly-p-phenylene vinylene derivative (BEH-PPV) has been used together with poly(9,9-dioctylfluorene) (PFO). BEH-PPV has a highest occupied molecular orbit (HOMO) of 5.3 eV.⁷ Values for the HOMO of PFO range from 5.8 eV⁸ to 6.1 eV⁹. Thus the interface energy barrier between BEH-PPV and PFO is between 0.7 and 1.0 eV. It is expected that for such a large interface energy barrier the current will be limited by the injection rate.

2 Experimental

The devices that have been investigated are of three types, they are all designed to measure the hole transport eq injection through the polymer layers. For that reason, all the devices have a top contact of gold. Gold has a high work-function, and will therefore block the electron injection. First of all, devices have been constructed where a layer of BEH-PPV has been spin-coated on top of an Ohmic indium tin oxide (ITO) bottom contact. Furthermore, devices have been constructed where the ITO has been covered by the BEH-PPV, and on top of this layer PFO has been spin-coated. Via thickness measurements it has been confirmed that the total thickness equals the thickness of the two separate layers

(BEH-PPV and PFO), prepared under the same spin-coat conditions. The third device type consist of a bottom contact of Pt, on top of which the PFO has been spin-coated.

3 Results

In figure 1 the current density voltage (J - V) characteristic of the ITO/BEH-PPV/Au device has been shown, together with the ITO/BEH-PPV/PFO/Au device. It is demonstrated that the current through the ITO/PPV/Au device is space charge limited (SCL). The mobility amounts to $\mu_0=1.5 \times 10^{-10} \text{ m}^2/\text{Vs}$ for low field, whereas the field dependence can be described by $\mu_0 \times \exp(\gamma\sqrt{E})$, with $\gamma=3 \times 10^{-4} (\text{m/V})^{1/2}$.

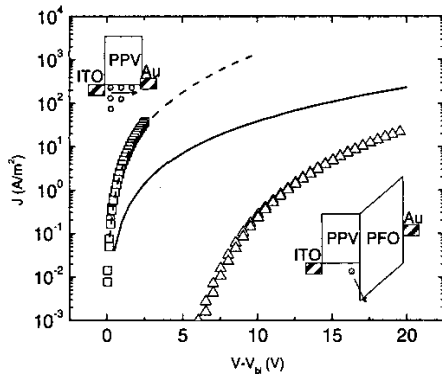


Figure 1, current density as a function of voltage, for two different devices. The ITO/PPV/Au device has a thickness of $d=140 \text{ nm}$, whereas the ITO/PPV/PFO/Au device has a total thickness of $d=370 \text{ nm}$, the PFO layer being $d=230 \text{ nm}$ thick. The dashed line shows the calculated SCL current through the ITO/PPV/Au device, the solid line represents the calculated SCL current through the ITO/PPV/PFO/Au device.

It is observed that the ITO/PPV/PFO/Au device indeed carries a much lower current. In order to discriminate between a SCL current for this thick device, or an injection limited (IL) current due to the hole barrier at the PPV/PFO interface, a device model has been used to calculate the SCL current in such a double layer device without interface barrier. The device model is similar to that for a single layer,¹ where the physical parameters for both layers have been included. The calculated SCL current for such a device without interface barrier is also plotted in Figure 1 as a solid line. It is observed that the measured ITO/PPV/PFO/Au device current (squares) is indeed much lower.

To demonstrate the IL nature of the current through the ITO/PPV/PFO/Au, the current has been plotted as a function of electric field for different thicknesses. The JV characteristics should fall on top of each other, as an IL current is a function of electric field $J=J(E)$. It is shown in figure 2 that the current as a function of electric field does coincide for the different thicknesses, which

confirms the IL nature of the current through the device, in correspondence with the large injection barrier of PPV/PFO of $\phi_b=0.7 \text{ eV}$.

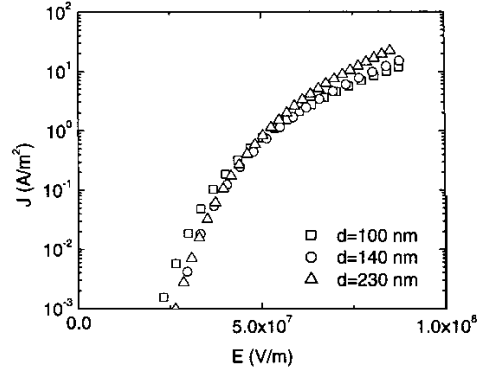


Figure 2, current density as a function of electric field for three different device thickness at room temperature.

Now we are sure about the IL nature of the current through ITO/PPV/PFO/Au, one might ask whether the injection current indeed differs from the injection current over a MOI. For this reason Pt/PFO/Au devices have been made. It has been measured with Kelvin probe technique¹⁰ that the work-function of Pt under ambient conditions amounts to $\sim 5 \text{ eV}$, which is much lower than the value in vacuum. This has also been observed for other metal electrodes.¹¹ The energy barrier for Pt/PFO is therefore comparable or even larger as for the PPV/PFO system.

In figure 3, the J - E characteristics of both a ITO/PPV/PFO/Au and a Pt/PFO/Au device are shown, both for a thickness of $d=160 \text{ nm}$ of the PFO layer. It is observed that for low voltages the leakage current in the Pt device dominates, while for the ITO/PPV/PFO/Au device the device current no leakage is observed. This demonstrates the superiority in device performance for a polymer bottom contact layer with respect to metal contacts.

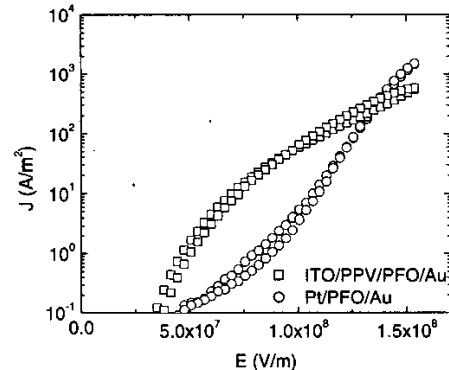


Figure 3, current density as a function of electric field for two different device types, a MOI and a OOI.

Furthermore, it is observed from Figure 3 that the IL current of the ITO/PPV/PFO/Au device (OOI) increases strongly with electric field at low applied biases, whereas the IL current through the Pt device (MOI) only comes up at much higher fields. The current in the OOI is larger at not too high electric field, which is an indication that the injection barrier for the Pt/PFO contact is indeed larger compared with the OOI.

It is also observed that at high fields the field dependence of the IL current of the MOI is larger. This indicates that for the MOI the image force lowering is present, which reduces the injection barrier for increasing electric field. The strong increase of the injection current at low fields for the OOI is not yet clear and will be subject to further study.

3. Conclusion

The current in a double layer device with a large interface barrier ($\phi_b=0.7-1.0$ eV) is strongly reduced with respect to the space charge limited current for such a device. It is found that the current-density scales with electric field, indicating that the current across the OOI is injection limited. Moreover, it is found that the current density voltage characteristics are completely different for an OOI and a MOI, indicating the different nature of the organic-organic and the metal-organic contact. The strong rise with electric field for a MOI can be explained by the image force lowering, while the difference in injection efficiency at moderate fields can be attributed to the absence of image force in the OOI, resulting in a smaller back-flow of carriers.

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