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Charge injection in organic light-emitting diodes: Tunneling into low mobility materials

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We present device model calculations for the current–voltage (I–V) characteristics of organic diodes and compare them with measurements of structures fabricated using MEH-PPV. The measured I–V characteristics have a Fowler–Nordheim (FN) functional form, but are more than three orders of magnitude smaller than the calculated FN tunneling current. We find that the low mobility of the organic materials leads to a large backflow of injected carriers into the injecting contact. These results account for the experimental observations and also demonstrate how transport layers in multilayer organic light-emitting diodes can be used to improve carrier injection. [S0003-6951(96)02341-8]

Organic light-emitting diodes (LEDs) have potential fabrication, mechanical, and cost advantages.[1–3] There has been significant progress both in the performance of these devices and in understanding the device physics governing their operation.[4] Because of these advances, organic light-emitting diodes are being seriously considered for large area applications including alphanumeric and flat panel displays.

A recent, detailed investigation of single carrier devices (i.e., devices in which only one of the contacts can inject carriers efficiently) has shown that for devices with fairly large Schottky energy barriers the current–voltage characteristics have a Fowler–Nordheim (FN) tunneling functional form at large forward bias[5]

$$J_{F+N}=J_0\left(\frac{E}{E_0}\right)^2 e-\left(\frac{E_0}{E}\right). \tag{1}$$

Here, J_{F+N} is the tunneling current, E is the field at the contact, and the characteristic current J_0 and field E_0 are given in terms of the Schottky energy barrier.[5] In Ref. 5, devices of differing length were considered and the measured current as a function of field was found to be independent of the device length as expected when the current flow is limited by tunneling injection. However the absolute magnitude of the measured current was over three orders of magnitude smaller than given by the FN tunneling expression [Eq. (1)]. These results show that tunneling is the dominant injection mechanism at high bias and large Schottky energy barrier, but that an additional process must be included to understand the device operation. Band bending[6] and image force[7] modifications to the tunneling current do not reduce the injected current by the necessary three orders of magnitude to account for the measured results.

Here, we present device model calculations for single carrier organic diodes and compare them with measurements of structures fabricated using the electroluminescent conjugated polymer poly[2-methoxy, 5-(2′-ethyl-hexyloxy)-1,4-phenylene vinylene] (MEH-PPV). We find that, due to the low mobility typical of organic materials, there is a large backflow of injected carriers into the injecting contact. The backflow current is the time reversed process of thermionic emission, that is, carriers on the organic side of a metal/organic contact fall back into the metal. It is a well known process in inorganic semiconductor Schottky diodes and at thermal equilibrium exactly cancels the thermionic emission current such that there is no net device current.[8,9] At high bias, a combination of FN tunneling injection and backflow currents establishes a carrier density near the injecting contact. The device current results primarily from drift of these carriers away from the injecting contact. The net device current is much smaller than either the tunneling injection or the backflow current, which nearly cancel. As a result, the net device current has nearly the FN functional form but is several orders of magnitude smaller than the tunneling injection current in absolute magnitude. This result accounts for the observations in Ref. 5.

The device model consists of drift-diffusion equations for electrons and holes together with Poisson's equation for the electrostatic potential. Charge injection at the metal/organic interface is described by thermionic emission (dominant at low bias) and FN tunneling (dominant at high bias). The net current at the contact is the injected current minus the backflow current. The backflow current is proportional to the carrier density at the contact. The proportionality constant is determined by detailed balance at thermal equilibrium. The equations are spatially discretized using the Scharfetter and Gummel scheme.[10] A voltage ramp is applied and the time dependent equations are integrated forward in time, starting from thermal equilibrium until steady state is reached. To describe single carrier devices, the Schottky energy barrier for one of the carriers is taken to be too large for effective injection.

Figure 1 shows a comparison of the measured and calculated I–V characteristics for a 120 nm Al/MEH-PPV/ITO hole-only device structure. ITO is a good hole injecting contact for MEH-PPV, but the energy barrier for electron injection from the Al contact is too large for efficient injection. The inset of Fig. 1 shows a FN plot of the measured and calculated currents. Two parameters are required for the one carrier device model, the Schottky energy barrier and the mobility. The Schottky energy barrier for hole injection at

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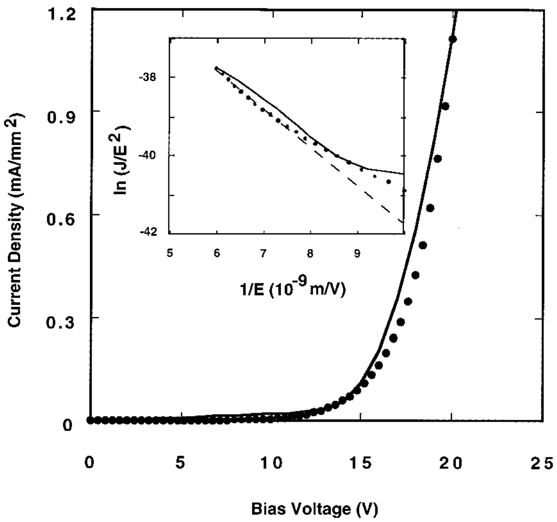

FIG. 1. The calculated (solid line) and measured (dots) I–V characteristics for an Al/MEH-PPV/ITO diode. The inset shows a Fowler–Nordheim plot of the same results.

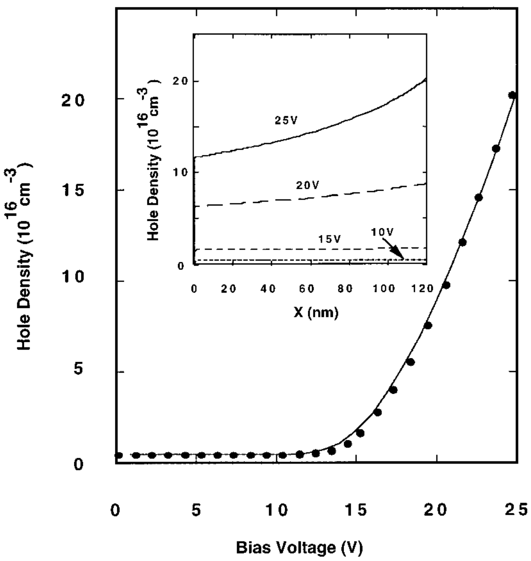

FIG. 2. The calculated hole density at the ITO contact for the Al/MEH-PPV/ITO device as a function of the applied bias. The dots are the scaled Fowler-Nordheim current. The inset shows the hole density profiles throughout the device for different biases. The ITO (hole injecting) contact is on the right side ($X = 120$ nm).

the ITO/MEH-PPV interface is taken to be 0.34 eV and the hole mobility is taken to be 8.5×10^{-6} cm^2/V s. Tunneling dominates injection for biases larger than about 14 V. The fit to the data is good, and as shown in the inset, both measured and calculated currents satisfy the FN functional form for biases at which tunneling dominates injection. Devices of differing length were also considered and the calculated current as a function of field was found to be independent of the device length, consistent with the experimental results of Ref. 5.

The hole density near the hole injecting contact is determined by a competition between injection and backflow. Figure 2 shows the calculated hole density at the hole injecting contact as a function of bias. The inset shows the calculated hole density as a function of position for several values of the bias voltage. At low bias, where thermionic emission dominates injection, the hole density at the contact is essentially constant and given by the equilibrium value. At high bias, where tunneling dominates injection, the hole density is proportional to the tunneling injection current and has the FN functional form. As also shown in the inset, the hole density is a slowly varying function of position and thus diffusion currents are small. [The hole (electron) injecting contact is at the right (left) in the inset.]

The steady state device current is constant across the structure, and for a hole-only device can be written as the difference between the injection and backflow current at the hole injecting contact. The device current is given by

$$(J_{\mathrm{FN}}+J_{\mathrm{TH}})-J_{\mathrm{BF}}=J_D, \tag{2}$$

where the total injected current ($J_{\mathrm{FN}}+J_{\mathrm{TH}}$) is the sum of the FN tunnel and thermionic currents, J_{BF} is the backflow current, and J_D is the device current. The backflow current is given by

$$J_{\mathrm{BF}}=\nu P(L), \tag{3}$$

where $P(L)$ is the hole density at the contact and the proportionality constant ν is determined by Eq. (2) at equilibrium where J_{FN} and J_D vanish. Because the diffusion current at the hole injecting contact is small, the device current is well approximated as

$$J_D=e\mu E(L)P(L), \tag{4}$$

where μ is the hole mobility and $E(L)$ is the electric field at the contact. $P(L)$ can be found using Eqs. (2)–(4) and at high bias the ratio of tunneling injection current to total current becomes

$$\frac{J_{\mathrm{FN}}}{J_D}=\frac{\nu+e\mu E(L)}{e\mu E(L)}. \tag{5}$$

Due to the low mobility of organic materials, $e\mu E(L)$ is much smaller than v and the device current is much smaller than the tunneling injection current. The ratio of the right-hand side of Eq. (5) is a slowly varying function of bias compared to the exponential dependence of J_{FN}. Therefore, J_D has nearly the FN functional form, but is orders of magnitude smaller than the tunneling current when tunneling dominates injection. Figure 3 shows a plot of the calculated ratio J_{FM}/J_D as a function of bias. Above a bias of about 14 V, where tunneling dominates injection, the ratio is approximately constant with a value of about 5000. The inset of Fig. 3 shows the electric field as a function of position. The field is slowly varying across the device.

Due to the low mobility, the device current is limited by drift of the hole density away from the hole injecting contact. At high bias this hole density is determined by a competition between tunneling injection and backflow. The addition of a transport layer at the contact with a larger mobility but also a larger Schottky energy barrier can increase the net current flow by increasing the drift current for a given hole density. Of course, the larger Schottky barrier reduces the hole density at the interface so there is a trade-off between a higher mobility and a larger Schottky energy barrier. Figure 4 com-

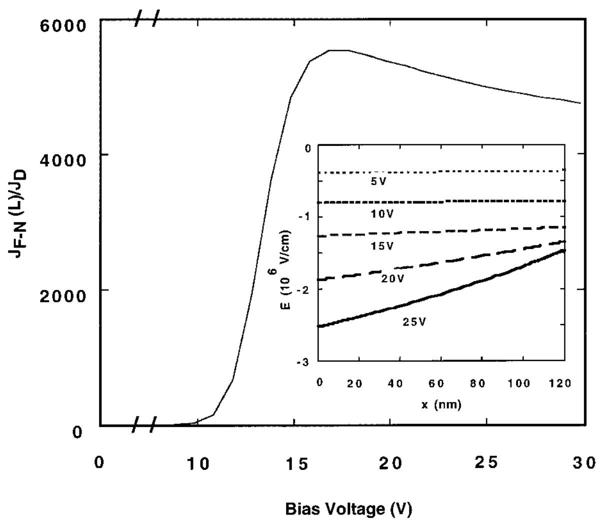

FIG. 3. The ratio of the FN injected tunneling current at the ITO contact to the device current as a function of bias. The inset shows the electric field as a function of position for different biases.

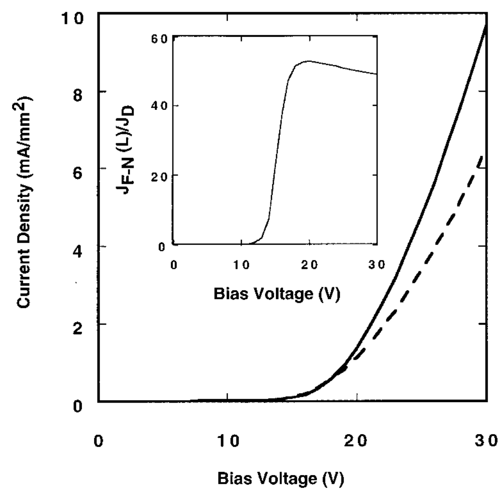

FIG. 4. Comparison of the $I-V$ characteristics for a single layered Al/MEH-PPV/ITO device (dashed line) and a bilayered Al/MEH-PPV/transport-layer/ITO device (solid line). The inset shows the ratio of the FN injected tunneling current at the ITO contact to the device current as a function of bias.

pares the calculated currents as a function of bias for the 120 nm single layer structure of Fig. 1 with a bilayered structure consisting of a 100 nm luminescent layer with the same properties as in Fig. 1 and a 20 nm hole transport layer with a 0.1 eV larger Schottky energy barrier and a mobility 100 times larger than the luminescent layer. For these parameters the bilayered structure has a larger net current at a given bias voltage compared to the single layer structure even though the Schottky energy barrier of the hole transport layer is larger than the luminescent layer. The inset of Fig. 4 shows the ratio (J_{FN}/J_D) for the bilayered structure as a function of bias. As for the single layer structure, shown in Fig. 3, the ratio is approximately constant when tunneling dominates injection. Comparison with Fig. 3 shows that the value of the current ratio is reduced by approximately the mobility ratio. These results show how a larger mobility in a transport layer can reduce the backflow current and increase the total device current. The larger mobility of the transport layer can help compensate for the larger Schottky energy barrier to charge injection so that the benefits of blocking carriers from traversing the device by the transport layer can be realized without severely reducing injection.

In summary, we presented device model calculations for single carrier organic diodes and compared with them measurements of structures fabricated using MEH-PPV. The results account for the observations of Ref. 5 and show how transport layers in multilayer organic light-emitting diodes can be used to improve carrier injection.

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