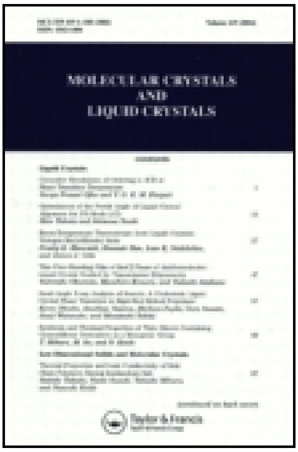

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Current-Voltage Characteristics of Polymer Light-Emitting Diode at Low Voltages

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CURRENT-VOLTAGE CHARACTERISTICS OF POLYMER LIGHT-EMITTING DIODE AT LOW VOLTAGES

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Abstract

We argue that the passage of current through a polymer light-emitting diode at low applied voltages $U < 10V$ is dominated by a sequential tunneling of electrons (holes) from a contact into the states of the conduction (valence) band via the chains of localized states in the gap. The current-voltage characteristics I(U), determined by such a mechanism, is predicted to be $|\ln I| \alpha 1/\sqrt{U}$.

The results of recent experiments on poly(paraphenylene venylene) (PPV) light-emitting diodes suggest that high enough applied voltages, $U \sim 10V$, the passage of current is dominated by tunneling of electrons (holes) from a metallic contact into the states of the conduction (valence) band. This situation is illustrated in Fig. 1 for the case of electron tunneling.

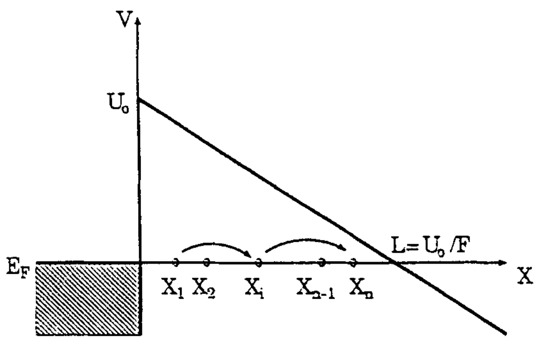

FIGURE 1 Schematic band profile in the conduction band of the device.

The magnitude of current is determined by the probability of tunneling. Let F be an applied electric field, F=U/D, D being the thickness of the device. Then the potential profile in the vicinity of a contact has the form $V(x)=U_0-Fx$, where U_0 is the barrier height, determined by the difference in the work functions between the contact and the conduction band of the polymer. The tunneling integral for such a triangular barrier determines the form of the current voltage characteristics $I \alpha \exp(-Q)$, with

$$Q = \frac{2\sqrt{2m}}{\hbar} \int_0^L dx\sqrt{V(x)} \tag{1}$$

where $L = U_0/F$. Calculating the integral, we reproduce the classical Fowler-Nordheim result

$$Q = \frac{4}{3}\frac{\sqrt{2m}}{\hbar}\frac{U_0^{3/2}}{F} = Q_0 \tag{2}$$

The experimental current-voltage characteristics[1,2] were fitted by the dependence $|\ln I| \;\alpha\; F^{-1} \;\alpha\; U^{-1}$ and the agreement seems reasonable for the high-voltage range $U \sim 10V$. Let us estimate the corresponding tunneling length $L = U_{0/F} = U_0D/U$. Assuming $U_0 \sim 0.5V$, D = 1000 Å and $U \sim 10V$ we get $L \sim 50$Å. For lower voltages this width is much larger, so that the direct tunneling becomes impossible. The experiments, however, show that at voltages much less than 10V the current is nonzero and its magnitude is much higher than that predicted by Eq.(2) (excess current).

In the present paper we suggest that the physical mechanism for the excess current is the sequential tunneling of electrons via the localized states in the gap. To illustrate this mechanism assume that there is only a single state, i, on the way of tunneling electron (see Fig. 1). Then the process of passage of an electron through the barrier consists of two steps: i) tunneling from the contact to the localized state and ii) tunneling from the localized state into the conduction band. Let us denote with τ_{ci} and τ_{ib} the corresponding waiting times. We assume that due to strong coupling of localized state to the phonons the electron phase is completely lost after tunneling, so that there is no coherence in the processes $c \to i$ and $i \to b$. The average occupation number of the localized state is determined by the condition that the average flow of electrons on the first step (c→i) is equal to the average flow on the second step (i→b) and equal to the total current[3]

$$\frac{1-\langle n_i\rangle}{\tau_{ci}} = \frac{\langle n_i\rangle}{\tau_{ib}} = \frac{I}{e} \tag{3}$$

The solution of the system is easily found to be

$$\langle n_i\rangle = \frac{\tau_{ib}}{\tau_{ci}+\tau_{ib}}, \quad I = \frac{e}{\tau_{ci}+\tau_{ib}} \tag{4}$$

The times τ_{ci} and τ_{ib} can be expressed through tunneling integrals c→i and i→b as follows

$$\tau_{ci} = \tau_0 \exp Q_{ci}, \quad \tau_{ib} = \tau_0 \exp Q_{ib}, \tag{5}$$

where

$$Q_{ci} = \frac{2\sqrt{2m}}{\hbar}\int_0^{x_i} dx\sqrt{U_0-Fx} = Q_0\left[1-\left(1-\frac{Fx_i}{U_0}\right)^{3/2}\right]$$
$$Q_{ib} = \frac{2\sqrt{2}\,m}{\hbar}\int_{x_i}^{L} dx\sqrt{U_0-Fx} = Q_0\left(1-\frac{Fx_i}{U_0}\right)^{3/2} \tag{6}$$

We see that the product $\tau_{ci}\tau_{ib}=\tau_0^2 \exp(-Q_0)$ does not depend on the position x_i of the localized state in the barrier. Since both times depend on x_i exponentially, the sum $\tau_{ci} + \tau_{ib}$, as a function of x_i, has a sharp minimum, determined by the condition $Q_{ci}(x_i) = Q_{ib}(x_i)$. Using Eq. (6) we obtain

$$x_i = (1-2^{-2/3})L \tag{7}$$

For the maximal value of current caused by a tunneling via a single localized state we get

$$|\ln I| = \frac{Q_0}{2} \tag{8}$$

We see that the exponent in Eq. 8 is twice as small as in contribution from the direct tunneling.. This exponent results from the configurations for which $\tau_{ci} = \tau_{ib}$. However, the contribution from the sequential tunneling contains a small prefactor, p, which is the probability to find a localized state in the vicinity of the point x_i (7) and with energy close to the Fermi level.

The contribution to the current, I_n, from the sequential tunneling via the chains consisting of arbitrary number of localized states, N, can be found in the similar way[4]. The optimal positions of the localized states x_n, are determined by the condition that the waiting time for tunneling between the successive localized states n and n+1 is the same for all. By analogy to Eq. 5 we express this time as

$$\tau_n = \tau_0 \exp Q_n, \tag{9}$$

where

$$Q_n = \frac{2\sqrt{2m}}{\hbar}\int_{x_n}^{x_{n+1}} dx\sqrt{U_0 - Fx} = Q_0\left[\left(1-\frac{Fx_n}{U_0}\right)^{3/2} - \left(1-\frac{Fx_{n+1}}{U_0}\right)^{3/2}\right] \tag{10}$$

The condition that all Q_n are equal allows to find the optimal positions x_n

$$x_n = \left[1-\left(\frac{N+1-n}{N+1}\right)^{2/3}\right] L. \tag{11}$$

Then the exponent in the current through the chain is just $Q_0/(N+1)$ and the contribution to the total current from the chains with N localized states can be presented as

$$I_N = p^N \exp\left[-\frac{Q_0}{N+1}\right], \tag{12}$$

where p^N is the probability of the formation of the chain. Eq. 12 represents the product of two factors, one increasing and the other decreasing with N (since $p << 1$). The product is maximal for

$$N = N_{opt} = \left(\frac{Q_0}{\mathcal{L}}\right)^{1/2} - 1, \tag{13}$$

where $\mathcal{L} = \ln(1/p)$. Substituting (13) into (12) we get the final result for the current determined by a sequential tunneling

$$|\ln I| = \frac{2Q_0}{N+1} = 2\mathcal{L}^{1/2}Q_0^{1/2}. \tag{14}$$

Using (2) the current-voltage characteristics can be presented as

$$|\ln I| = \frac{4\mathcal{L}^{1/2}}{3^{1/2}}\left(\frac{2md^2}{\hbar^2}\right)^{1/4}\frac{U_0^{3/4}}{U^{1/2}}. \tag{15}$$

Eq. (15) is the main result of the present paper. We see that the mechanism considered results in the slower voltage dependence of current, $|\ln I| \propto 1/\sqrt{U}$, than it is for direct tunneling. The important feature of the result obtained is its rather weak sensitivity to the concentration of localized states in the gap, which is usually unknown. Note also a rather weak dependence of the result on the electron mass, the value of which is not established unambiguously by now.

In Fig. 2 we show two experimental current-voltage characteristics of light-emitting diodes. The first one (Fig. 2a) is the data from the paper[2] plotted in the axes lnI vs $F^{-1/2}$. The second one (Fig. 2b) was measured by us on the structure: ITO-PPVMEH-Ca with the thickness of the polymer $D \approx 1200$Å. The data was taken using HP4145B semiconductor parameter analyzer and plotted in the axes ln I vs $U^{-1/2}$. We see that in agreement with prediction of the theory developed the curves are linear within a rather wide interval of applied voltages (electric fields).

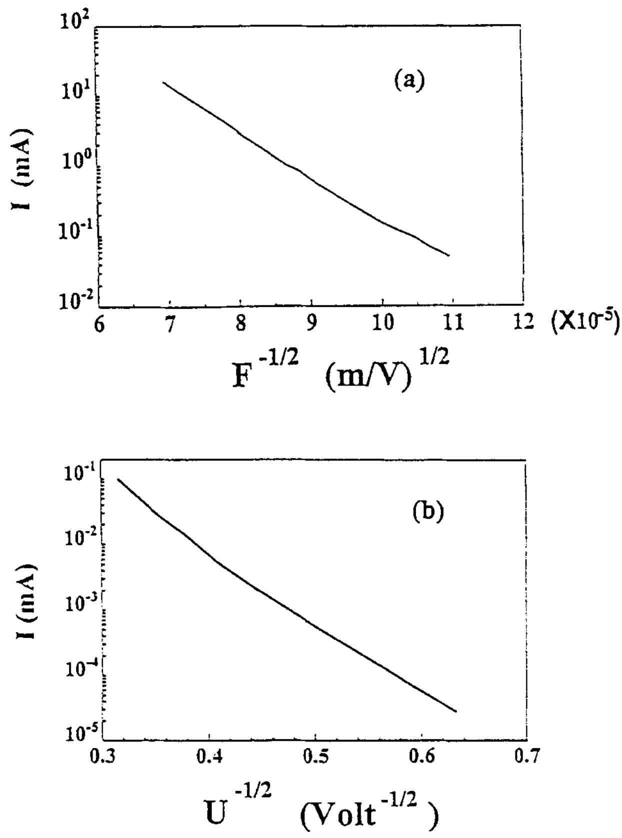

FIGURE 2 Experimental current-voltage characteristics of two light-emitting diodes. (a) The data[2] is plotted in the axes lnI vs $F^{-1/2}$. (b) Our data taken on the structure ITO-PPVMEH-Ca.

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