Proceedings of the IV International Seminar on Organic Materials for Molecular Electronics, Zajączkowo 1994

ELECTRIC CURRENT FLOW THROUGH TETRACENE LAYER-ANTHRACENE CRYSTAL JUNCTION

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Theoretical and experimental analysis was carried out on electric currents limited by the potential barrier governing the flow of holes from the tetracene layer to the anthracene crystal. Theoretical spatial distributions of charge carriers near the barrier were determined, as well as current-field dependences for the currents flowing through the investigated junction in the presence and in the absence of illumination. A current-field characteristic of the junction conditioned current is described by dependence of the $j \propto E_0^3$ type in the presence of illumination and by the $j \propto E_0^{2l+1}$ type in the lack of illumination, where *l* is the characteristic parameter of the trap distribution. Experimental research of the hole currents flowing through the polycrystalline tetracene layer-anthracene monocrystal junction confirmed the theoretical predictions about the current-field characteristics. A remarkably unequivocal confirmation was obtained for the current-field dependences in the presence of illumination.

PACS numbers: 72.40.+w

1. Introduction

A junction found in systems of two or more different organic materials is an interesting subject of research for physics of organic materials and molecular electronics. It is connected, *inter alia*, with the search for efficient light emitting diodes [1-3] and organic photovoltaic cells [4-6].

Discontinuity of energy bands occurring on the junction may also lead to a major asymmetry in the system's current-voltage characteristics. Such a rectifying junction between two different phthalocyanines was examined in Ref. [7]. The current paper presents our analysis of electrical properties of the anthracene-tetracene (A-T) system, which with the well-known properties of both materials may serve as a model molecular junction. The objective of this paper is to determine the spatial distribution of charge carriers in the charge-accumulating layer (i.e. in tetracene) and the current-field characteristics of the illuminated and the unilluminated junction with reverse polarization. It is a continuation of our earlier research [8-11].

2. Experimental basis

An anthracene monocrystal (A)-vacuum-evaporated tetracene layer (T) junction has been examined, with electrodes well injecting holes (CuI [12]), polarized in the reverse direction (+ tetracene, see Fig. 1). The difference in ionization energy between anthracene and tetracene equals $W_0 = 0.4$ eV [13] and this value may also be expected to be the potential barrier for holes flowing from tetracene to anthracene [11]. Radiation illuminating the system is transmitted through the anthracene crystal and absorbed in the tetracene layer. Actually, the tetracene layer is much thinner than anthracene ($b \ll a$).



Fig. 1. Schematic presentation of the anthracene-tetracene junction, I_c^{T} , I_c^{A} — ionization energy of tetracene and anthracene, respectively. Further explanation of the symbols in the text.

3. Theoretical calculation

3.1. Spatial distribution of electric field intensity and hole concentration in tetracene layer

To determine the spatial distributions of electric field intensity $E^{T}(x)$ and concentration of holes, trapped $p_{t}^{T}(x)$, and untrapped $p_{f}^{T}(x)$, we use the following current density equation:

$$j = e\mu^{\mathrm{T}} E^{\mathrm{T}}(x) p_{\mathrm{f}}^{\mathrm{T}}(x) - \mu^{\mathrm{T}} k T \frac{\mathrm{d} p_{\mathrm{f}}^{\mathrm{T}}(x)}{\mathrm{d} x} \approx 0, \qquad (1)$$

limited to the case of weak currents $(j \approx 0)$, and the Poisson equation

$$\frac{\mathrm{d}E^{\mathrm{T}}(x)}{\mathrm{d}x} = \frac{e\left[p_{\mathrm{t}}^{\mathrm{T}}(x) + p_{\mathrm{f}}^{\mathrm{T}}(x)\right]}{\varepsilon_{0}\varepsilon^{\mathrm{T}}} \approx \frac{ep_{\mathrm{t}}^{\mathrm{T}}(x)}{\varepsilon_{0}\varepsilon^{\mathrm{T}}}.$$
(2)

These are complemented with an expression describing the processes of charge carrier trapping and detrapping. For exponential energy distribution of traps $h(E) = (H/kTl) \exp(-E/kTl)$, where E is a trap depth, l — the characteristic parameter of distribution, and H — total concentration of traps, this expression takes the following form when optical detrapping prevails:

$$\frac{p_{\rm f}^{\rm T}(x)}{p_{\rm t}^{\rm T}} = \Theta = \frac{N_{\rm ef} l \sin(\pi/l)}{\pi H} \left(\frac{\xi \tau \gamma_{\rm h} \kappa I_0}{\nu}\right)^{1-1/l},\tag{3}$$

or the following form when thermal detrapping is dominant [14]:

$$p_{\rm f}^{\rm T} = N_{\rm ef} \left[\frac{l \sin(\pi/l)}{\pi H} \right]^l \left[p_{\rm t}^{\rm T}(x) \right]^l = D \left[p_{\rm t}^{\rm T}(x) \right]^l.$$

$$\tag{4}$$

In expression (3), true for $\kappa x \ll 1$, κ stands for the absorption coefficient, I_0 light intensity at the junction (x = 0), ξ — detrapping efficiency, τ — the lifetime of exciton, γ_h — the exciton trapped hole interaction constant. $N_{\rm ef}$ is the effective density of states and ν — the frequency factor. Equations (1), (2) and (3) or (4), taken with the boundary conditions

$$E^T(x=0) = E_0 \quad \text{and} \quad E(x \to -\infty) = 0,$$
(5)

enable us to obtain the required spatial distributions of electric field intensity and charge concentration for the illuminated system:

$$E^{\rm T}(x) = \frac{E_0}{1 - eE_0 x/2kT},\tag{6}$$

$$p_{\rm f}^{\rm T}(x) = \frac{\varepsilon_0 \varepsilon}{2kT} \left(\frac{E_0}{1 - eE_0 x/2kT} \right)^2,\tag{7}$$

and for the unilluminated system:

$$E^{\mathrm{T}}(x) = \frac{E_0}{1 - eE_0 x/2kTl},$$
(8)

$$p_{\rm f}^{\rm T}(x) = \left(\frac{\varepsilon_0 \varepsilon^{\rm T}}{2kTl}\right)^l D\left(\frac{E_0}{1 - eE_0 x/2kTl}\right)^{2l}.$$
(9)

The respective concentrations of trapped carriers can be determined from the formula (3) or (4). The above expressions describe the spatial distributions of the charge carriers in the presence of an external electric field in contradistinction to the systems Letermined by the carrier diffusion only [15].

3.2. Current-field characteristics

To determine the current-field characteristics we assume that there is no space charge in anthracene and that current density is adequately described by

$$j = e\mu_{\rm A} p_{\rm f}^{\rm A} E^{\rm A},\tag{10}$$

where

$$E^{\mathbf{A}} = \text{const} = E_0 \approx \frac{U}{a},$$
 (11)

and U is the external voltage. Unless major departures of the thermodynamic equilibrium occur, i.e. for moderate fields, hole concentrations in anthracene and tetracene are related as follows:

$$p_{\rm f}^{\rm A} = p_{\rm f}^{\rm T}(x=0) \exp\left(-\frac{W_0}{kT}\right). \tag{12}$$

Emploing expressions (7) and (9) for x = 0, we obtain the following characteristics:

$$j = e\mu^{A}\Theta \frac{\varepsilon_{0}\varepsilon^{T}}{2kT} \exp\left(-\frac{W_{0}}{kT}\right) E_{0}^{3} \propto U^{3}$$
(13)

for the illuminated system and

$$j = e\mu^{A} D\left(\frac{\varepsilon_{0}\varepsilon^{T}}{2kTl}\right)^{l} \exp\left(-\frac{W_{0}}{kT}\right) E_{0}^{2l+1} \propto U^{2l+1}$$
(14)

for the unilluminated system.

4. Experimental results

Exemplary results of measured current-field dependences of a CuI-A-T-CuI system are shown in Fig. 2.

These dependences can be approximated into the function $j \propto E_0^m$, where m = 3.2 for the illuminated system and m = 4.7 for the unilluminated system. The value of m = 3.2 is close to the theoretical value m = 3 (13). The parameter l = 1.8 (14) can be determined from the "dark" characteristic, which means that shallow traps, characteristic for molecular layers, prevail in the near junction region of tetracene. The obtained experimental results confirm our theoretical calculations.



Fig. 2. Electric current-electric field characteristics of the unilluminated (\bullet) and illuminated (\circ) anthracene-tetracene junction.

Acknowledgment

This work was supported in part by the project No. 204599101 of the State Committee for Scientific Research (Republic of Poland).

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