

Carrier Localization in Organic Transistors with Polymer Gate Insulators

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Abstract

A carrier localization model in organic field-effect transistors is discussed, based on the dipole disorder in polymer gate insulators. We first formulate the potential fluctuation due to polar groups of polymer gate insulators, and investigate the spatial distribution of electronic wavefunctions in the presence of the fluctuation by numerical calculation. The electronic states are found weakly localized over a few or a few tens of molecules in the most popular organic semiconductor, pentacene. The weakly-localized states are also probed experimentally by electron spin resonance (ESR), whose results are consistent quantitatively with the calculations.

1. Introduction

Organic field-effect transistors (OFETs) have attracted considerable attention because of their high potential for flexible and printable electronic devices. It has been believed that the best performance of OFETs can be achieved in the form of the perfect single crystal channels without any defects or grain boundaries [1,2]. However, since the charge transport occurs almost at the organic semiconductor/insulator interfaces, the charge transport should be also affected by the interaction between carriers and gate insulators. One example is the dielectric polarization effects discussed by Stassen *et al.*, while they did not take into account the spatial disorder of the gate insulators [3]. Here we show that the polar groups of polymer gate insulators should cause significant potential fluctuation at their surface, whose amplitude can be formulated analytically in our model. Its impact on the carrier localization is investigated by numerical calculations under the tight-binding approximation (TBA). We also compare the calculated results with our electron spin resonance (ESR) experiments.

2. Results and Discussions

We selected eight kinds of polymer insulators which are commonly used for organic transistors: polyvinyl alcohol (PVA), poly(chloro-p-xylylene) (parylene C) [4], poly(methyl methacrylate) (PMMA), poly(4-vinylphenol) (PVP), polyethylene oxide (PEO), CytopTM [5], polystyrene (PS), and poly(p-xylylene) (parylene N), whose molecular

structures are shown in Fig. 1. The dipole moment of each monomer unit was calculated by Gaussian 09 (MP2, 6-31G(d)) and listed in Table I. Large dipole moments more than 1 debye were obtained for PVA, parylene C, PMMA, PVP and PEO. By assuming that the dipoles are oriented at random and uncorrelated with each other, we formulated the amplitude of the potential fluctuation W as:

$$W = \frac{p}{\epsilon} \sqrt{\frac{n}{48\pi h}} \quad (1)$$

Here, p is the dipole moment, ϵ is the dielectric constant, n is the density of dipoles per unit volume, and h is the distance from the surface of insulating layer. According to the eq. (1), we calculated the amplitude of the potential fluctuation for a variety of polymer materials (Table I).

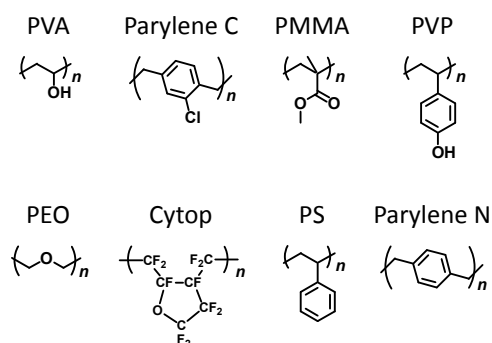


Fig. 1 Molecular structures of a variety of insulating polymers.

Table I Dipole moments and the amplitude of potential fluctuation for a variety of polymer insulators ($h = 0.8$ nm).

polymer insulator	dipole moment p (debye)	potential fluctuation W (meV)
PVA	1.9	130
parylene C	2.1	54
PMMA	1.8	52
PVP	1.6	41
PEO	1.3	25
Cytop	0.3	11
PS	0.2	7.7
parylene N	0.0	0.0

Since the amplitude of potential fluctuations is comparable to the transfer integrals for organic semiconductors, the electronic states near the band edge are expected to be localized by the Anderson localization (Fig. 2). To confirm this, we calculated the electronic states of pentacene film in the presence of the potential fluctuation, $W = 50$ meV, under the tight-binding approximation (TBA). The Hamiltonian matrix was diagonalized numerically, and all the eigenstates were obtained. Figure 3 shows the wavefunctions of ground, 2nd and 3rd states without and with potential fluctuation. The electronic states were found to be localized over a few pentacene molecules by the potential fluctuation. In such systems, the charge transport has to be assisted by thermal energy, by which carriers can be activated into the band states (in the case of multiple trap-and-release model) or can hop directly between the localized states (in the case of variable range hopping model).

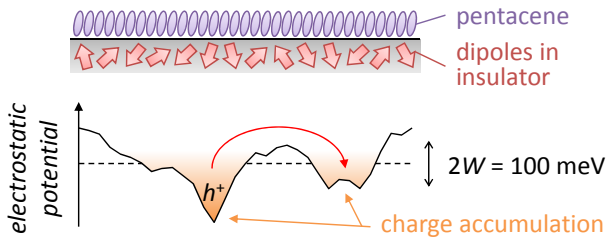


Fig. 2 Schematics of potential fluctuation at organic semiconductor/polymer insulator interface.

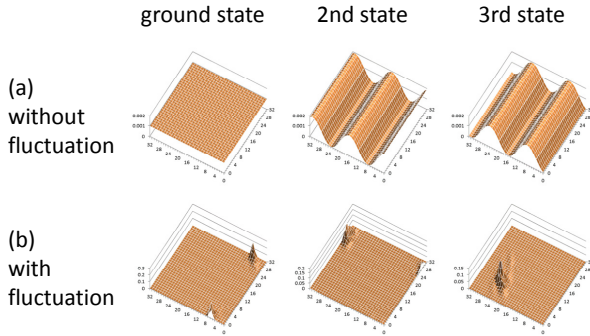


Fig. 3 Calculated wavefunctions (a) without and (b) with potential fluctuation.

Finally, we compare the calculated results with our electron spin resonance (ESR) experiments [6]. Figure 4 shows the distribution of the localization degree of electronic states obtained by ESR experiments and TBA calculations. Although we did not use any fitting parameters, the absolute scales of the two results are roughly consistent with each other, supporting the validity of the potential fluctuation model. The peak structure which is seen only in the experimental data is most probably due to point defects such as dislocations, vacancies or impurities.

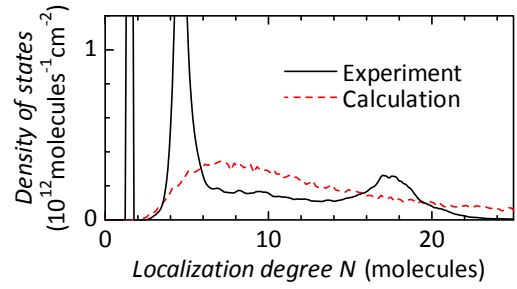


Fig. 4. Comparison between experimental and calculated density of states.

3. Conclusions

In summary, we formulated the potential fluctuation by dipole disorder at organic semiconductor/polymer insulator interfaces, and demonstrated its impact on the carrier localization by numerical calculation. The potential fluctuation should be a common feature for the interfaces between polymer insulators and a variety of semiconductor materials including graphene and oxide semiconductors.

References

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