Interpretation of trap-limited mobility in space-charge limited current in organic layers with exponential density of traps

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Charge carrier transport in disordered organic semiconductors, performed in electronic devices such as optoelectronic and photovoltaic ones, is usually affected by an exponential distribution of localized states in the band-gap (traps) under space-charge limited current. In this paper, we provide a full analysis for the trap-controlled transport of the single-carrier device in the frequency domain. Trap-limited mobility is interpreted in terms of the classical multiple-trapping picture with one transport state and the trapping-detrapping dynamics of the exponential density of traps. This allows us to provide a suitable explanation of the usual experimental features of the mobility dependence on voltage as along with the capacitance spectra. © *2011 American Institute of Physics*. [doi:10.1063/1.3622615]

I. INTRODUCTION

Over the last two decades, the application of light-emitting diodes (LEDs) to the technology of daily life has created an increasing interest, going beyond the traditional role of light indicators and displays to home appliances and general illumination.[1,2] At the same time, an exhaustive technological research effort was triggered by the potential applications of the new ongoing generation of these devices based on organic semiconductors.[3] Low-cost manufacturing processability and large area and flexible devices may be achieved by polymer-based organic light-emitting diodes.[4,5] Nevertheless, the performance of organic electronic devices strongly depends upon the charge transport process carried out, therefore, further understanding of the physical behavior of such materials is needed. For instance, the description of charge transport in organic layers by space-charge limited current (SCLC)[6–9] requires an interpretation of mobility by different semiempirical models (with field- or density-dependence), which is the crucial parameter governing the transport in the bulk.[10] Recently,[11] we have treated the relationship between the different models and in this paper we provide a full analysis of the interpretation of the capacitance and conductance spectra, along with a comparison with the observed experimental features.

In the 1990s, field-dependent mobility models were proposed by Bässler, as a result of assuming hopping transport in a Gaussian density of states (DOS).[12] Field-dependent mobility in organic layers, such as in sandwiched films composed of either poly(p-phenylene vinylene) (PPV) derivatives or aluminum hydroxyquinoline (Alq3), became widely accepted. However, Tanase *et al*. presented a comparison of mobility values for two solution-processed organic polymers: poly(2-methoxy-5-(3′,7′-dimethyloctyloxy)-p-phenylene vinylene) (OC_1C_{10}-PPV) and poly(3-hexyl thiophene) (P3HT), performed in two different configurations, i.e., field-effect transistors and hole-only diodes.[13] The first structure displayed mobility results up to three orders of magnitude higher than the latter configuration. These observations endorsed the density-dependent mobility model, proposed by Vissenberg and Matters in amorphous organic transistors, that stems from hopping percolation in an exponential DOS.[14] As demonstrated by Arkhipov *et al*., hopping transport in disordered materials can be reduced to a trap-controlled transport composed of an effective transport level and a broad distribution of localized states (traps) that only retain mobile charges.[15] Currently, several authors are considering this framework: transport via an extended state under the influence of an exponential density of traps.[11,16–18] In the present paper, we implement this assumption in SCLC to analyze its implications for charge carrier mobility.

From an experimental point of view, the determination of mobility is commonly given through the study of transit times (i.e., time needed for carriers to cross the sample electrode-to-electrode) in the wide range of the methods available in the literature: time-of-flight, transient electroluminescence, dark injection, and impedance spectroscopy (IS), among others.[19,20] The IS technique will focus our computational calculations to provide physical insights on the experimental measurements of capacitance spectra. As demonstrated in an earlier theoretical work of a single-carrier device in SCLC with only a single trap,[21] there is a strong correlation between the shape of the capacitance spectra and the nature of traps lying in the band-gap. In particular, a classification of them was established using its energy depth and its dynamic activity to capture and release charge carriers (i.e., shallow: fast and slow, and deep traps). Meanwhile fast-shallow traps were responsible for the delay of transit times (i.e., shifted capacitance step-ups), slow-shallow traps were for low-frequency capacitance increases. The aim of this paper is to extend these ideas to a wider set of traps, from a single to an exponential density in the band-gap,[11] in order to further test the theoretical framework with experimental capacitance data: a single-carrier device composed of N,N′-diphenyl-N,N′-bis(1-naphtylphenyl)-1,1′-biphenyl-4,4′-diamine (α-NPD).

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$$\tau_{trdc} = \frac{4}{3}\frac{L^2}{\mu(V_{bias} - V_{bi})}. \quad (26)$$

Inserting Eq. (25) into Eq. (26), it holds,

$$\mu = \frac{4}{3}\frac{L^2 f_{\max}}{0.72 \cdot (V_{bias} - V_{bi})}. \quad (27)$$

Calculations of mobility yielded mobility enhancement the more the voltage is applied in the bulk. In particular, the fitting to the classical field-dependent mobility expression,

$$\mu = \mu_0 \exp\left(\gamma\sqrt{F}\right), \quad (28)$$

with the approximation of $\sqrt{F} \approx \sqrt{V/L}$, provided experimental mobility values of $\mu_0 = 2.9 \times 10^{-4}\text{cm}^2/(\text{Vs})$ and $\gamma = 2.6 \times 10^{-3}(\text{cm/V})^{1/2}$. This mobility dependence upon voltage, measured by means of IS, is interpreted in the present paper as a trap-limited mobility governed by the dynamics of the fast-shallow traps in the band-gap. The field-dependent mobility is induced by the reduction of the trapping action as far as more voltage covers more trapping sites in the exponential distribution of localized-states. As regards the high frequency range (HF); it is composed of two different parts: a relatively wide plateau lying at the capacitance value of C_g, and a sharp decrease from a frequency cutoff onwards. The latter behavior of the HF is dominated by the series resistance of the whole device structure causing the capacitance drop.

In summary, the shape of the capacitance spectra with an exponential density of traps is strongly determined by the bias-voltage, providing: (1) a deviation of transit times translated into a field-dependent mobility, and (2) a low-frequency capacitance increase over the traditional $0.75C_g$ for trap-free materials.

IV. CONCLUSIONS

We have corroborated the theoretical framework of the multiple-trapping picture in organic layers that comprises an exponential density of trapping states under SCLC. The analysis of the capacitance spectra facilitates the interpretation of the voltage dependence of the trap-limited mobility and the low-frequency capacitance behavior. The presence of pure fast-shallow traps determines the limitation of the charge transport mobility, whereas slow-shallow traps cause the low-frequency capacitance to increase. Both features are modulated by two respective voltage-dependent energy regions in the band-gap.

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