A time-domain analysis of dipolar effects in copper phthalocyanine thin films on indium–tin–oxide substrates

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Abstract
Transient charging in response to pulsed bias of copper phthalocyanine (CuPc) organic thin films on indium–tin–oxide (ITO) was investigated by charge transient spectroscopy (QTS) in situ under ultra-high vacuum conditions. The charge relaxation phenomena in response to a bias pulse are modelled in terms of dipolar effects, considering alternatively possible orientation of the dipoles (polarization) under an applied external electric field and capture of charge carriers in dipole-induced electron states. Assuming a Gaussian energetic distribution of the density of states (DOS) as a consequence of disorder, a simple formalism is derived to determine the width (variance) \( \sigma_D \) of the DOS from QTS data. The isothermal QTS response of single-layer Ag/CuPc/ITO samples provides evidence for dipolar effects in CuPc, an example of using the new formalism in practice. The dipole moment of the CuPc molecule proved to be too small for the assignment of the amount of transient charges to the reorientation of dipoles. The model of trapping of charge carriers at a Gaussian DOS induced by polar molecules in a non-polar solid has mediated an estimate of the fraction of polar molecules from the experimentally assessed variance \( \sigma_D \) of the Gaussian DOS. The resulting dependence of \( \sigma_D \) on the dipole moment is in striking coincidence with literature data related to other types of polar molecules in molecular glasses.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The potential of thin organic layers as active components in electronic and opto-electronic devices such as unipolar transistors and organic light-emitting diodes (OLEDs) is attracting attention of both basic and applied research communities. Apart from attempts to dope organic materials, now it is well established that one deals with charge injection type of devices, the simplest ones being equipped with injecting contacts attached to at least partially disordered organic insulators. With reference to the profound review article by Brüting et al [1], situations can be met when it is possible to apply ‘classical’ approaches to the charge transfer through metal/organic interfaces, like Fowler–Nordheim tunneling or Richardson–Schottky thermionic emission. There is a common feature of many of the existing theories of charge injection to organics, namely the energy barrier for injection being determined by the difference between the work function of the (metal) electrode and the ionization energy or electron affinity of the organic dielectric. Regarding the role of traps in the injection phenomena, the more or less modified theory of injected steady-state trap-limited currents, found in the monograph by Lampert and Mark [2], is often applied [1].

A special category of organic substances is represented by π-conjugated small organic molecules. Charge redistribution as a consequence of electronic polarization, usually not considered in the case of inorganic semiconductors, plays an important role in modifying prominent energy levels of
individual molecules [3–5]. When trying to express the forward current–voltage (I–V) characteristics, the ‘classical’ approaches fail to explain the temperature dependence of the I–V curves. Prompted by this failure, new concepts emerged based on the presence of a distribution of hopping states in the dielectric [6]. The initial step is the injection of a charge carrier into a Gaussian distribution $g(E)$ of the density of states (DOS) with variance $\sigma_\text{DOS}$ centred around an energy $E_c$. Archipov et al [6] arrived at an analytical formula for the injection current of a single-layer device, including even a backward current component due to the image potential induced in the metal electrode. Later on, the applicability of the model was extended, covering also the case of charge carrier hopping at the interface separating two disordered organic materials, where recombination takes place [7]. One of the outcomes of the model is the finding that the leakage current of an OLED is determined by the rate of injection across the interface while the rate of radiative recombination being proportional to the first-jump rate. Finally, Baldo and Forrest [8] treated charge injection phenomena at metal/AIq3 interfaces taking into account the impact of true interfacial dipoles on adjacent molecules. The related DOS energy was supposed to be broadened to a Gaussian distribution by local disorder in the interfacial dipole field [8]. This is a situation similar to that encountered in the past when for a host non-polar matrix containing polar guest molecules (permanent dipoles) a simple relation between the Gaussian width $\sigma$ of the DOS and the relative concentration of polar (guest) molecules times their dipole moment was predicted and verified experimentally [9]. The electrostatic potential $\psi$ generated by the dipoles enters the probability density $g(\psi)$ as a direct reflection of the energy distribution $g(E)$ of hopping states in the system. When simulating short range order effects, Dieckmann et al [9] mentioned the possible effect of partial dipolar alignment. Yet, in order to simplify the modelling the dipoles were assumed as immobilized ones.

The present work aims at detecting charge relaxation in response to a sudden change of the applied electric field in a system for which alignment of true interface dipoles can be expected. This is an attempt to explore the dynamic response of the system after disturbing the equilibrium. Unlike the goals behind the studies cited above, we shall also investigate the kinetics of dipolar effects expressed in terms of the respective time constants. Effects of transient charging observed in the kinetics one would expect. This is an attempt to explore the dynamic response to a sudden change of the applied electric field in a system. When simulating short range order effects, Dieckmann et al [9] arrived at an analytical formula for the injection current of a single-layer device, including even a backward current component due to the image potential induced in the metal electrode. Later on, the applicability of the model was extended, covering also the case of charge carrier hopping at the interface separating two disordered organic materials, where recombination takes place [7]. One of the outcomes of the model is the finding that the leakage current of an OLED is determined by the rate of injection across the interface while the rate of radiative recombination being proportional to the first-jump rate. Finally, Baldo and Forrest [8] treated charge injection phenomena at metal/AIq3 interfaces taking into account the impact of true interfacial dipoles on adjacent molecules. The related DOS energy was supposed to be broadened to a Gaussian distribution by local disorder in the interfacial dipole field [8]. This is a situation similar to that encountered in the past when for a host non-polar matrix containing polar guest molecules (permanent dipoles) a simple relation between the Gaussian width $\sigma$ of the DOS and the relative concentration of polar (guest) molecules times their dipole moment was predicted and verified experimentally [9]. The electrostatic potential $\psi$ generated by the dipoles enters the probability density $g(\psi)$ as a direct reflection of the energy distribution $g(E)$ of hopping states in the system. When simulating short range order effects, Dieckmann et al [9] mentioned the possible effect of partial dipolar alignment. Yet, in order to simplify the modelling the dipoles were assumed as immobilized ones.

The ultra-high vacuum (UHV) facility used for organic beam deposition was described in our previous work [10]. Therefore, we concentrate on the description of fabricating Ag/CuPc/ITO samples analysed in the present study. ITO coated glass (PGO, 100 $\Omega$/square) was first cleaned by ultrasonic baths in acetone and isopropanol for 30 min each, followed by boiling in these solvents for 5 min. After drying under nitrogen flux, the substrates were transferred into the UHV chamber (base pressure better than $2 \times 10^{-10}$ mbar) where CuPc films were thermally evaporated using organic molecular beam deposition. CuPc of 99.96% purity purchased from SynTec was directly used without any further purification. The results shown here correspond to devices with thickness $L = 300$ nm of CuPc. Immediately after the organic deposition, 300 nm thick circular Ag dots were vapour deposited in the same chamber through a shadow mask ($\phi = 0.5$ mm, area $A = 1.96 \times 10^{-2}$ m$^2$). A complete electrical characterization (I–V, capacitance–voltage, charge transient spectroscopy—QTS) was conducted in situ at 300 K. The QTS instrumentation comprised a charge-to-voltage converter (integrator), three-gated integrators for exponential averaging and a mixer, the latter combining the outputs of the gated integrators according to the algorithm defined by equation (1) of section 3.

### 3. Isothermal QTS

Before presenting the analysis of experimental data on isothermal QTS, the capability of the method of providing the width $w$ of a Gaussian distribution of relaxation times $\tau$ is demonstrated. The biasing sequence is based on a bias pulse $U_0(t)$, of duration $\Theta_1$ superimposed on a quiescent dc bias $U_s$ applied to the bottom ITO electrode. Actually, there is a train of excitation pulses with a repetition period $\Theta_2 = \Theta_1 + \Theta_3$, enabling hardware supported averaging of each data point. The data processing during the rest ($\Theta_3$) of the repetition period consists of taking three samples of the transient charge $Q(t) = Q_0 \exp(-t/\tau)$ at respective delays $t_1, 2t_1$ and $4t_1$ after the trailing edge of the pulse, in order to obtain a weighted sum

$$\Delta Q(t_1, \tau) = Q(t_1) - \frac{3}{2}Q(2t_1) + \frac{1}{2}Q(4t_1).$$

where $Q_0$ stands for the full relaxing charge, the first-order (exponential) kinetics being characterized by the discrete time constant $\tau$:

$$\tau(T) = \tau_0 \exp \left( \frac{E_p}{kT} \right),$$

where $E_p$ is the thermal activation energy of the charge relaxation, $\tau_0$ is a pre-exponential factor, $k$ is the Boltzmann’s constant and $T$ is the temperature in K. The correlated charge $\Delta Q$ in equation (1) represents a filter of time constants $\tau$, its response peaking at $t_{\text{max}} \approx \tau$ with sufficient accuracy.

The practical implementation of an isothermal ($T = \text{const}$) QTS measurement takes the form of a scan of the time window $t_1$ within an interval as broad as possible. Here, the interval from $t_{\text{min}} = 2 \times 10^{-6}$ s to $t_{\text{max}} = 0.6$ s was employed, while taking $\Theta(t_1) = \Theta_1 + 4t_1$ and $\Theta_3 = 1$ ms. To exemplify the performance of the filter (1), figure 1 is included showing simulated QTS spectra for three charge relaxations differing in their respective time constants. An important point to make is that both the amplitude $\Delta Q_m$ and the half-width (FWHM $= 0.78$) are invariant to the parameters entering the right-hand side of equation (2). For the first-order $Q(t)$ kinetics one is expecting a simple relation between $\Delta Q_m$ and the total charge $Q_0$:

$$\Delta Q_m \approx 0.17Q_0.$$
The experimental quantity \( \Delta Q_m/0.17 \) is equal to the total relaxing charge \( Q_0 \) only if the condition \( \Theta_1 \geq 5\tau \) is fulfilled, taking the same \( \tau \) for both charging and discharging transients and writing \( Q_0 = Q_0[1-\exp(-\Theta_1/\tau)] \) for the charge acquired by the dielectric during the pulse. Strictly speaking, a finite duration \( \Theta_2 \) of the rest of the period has to be taken into account, as demonstrated by Barančok et al. [11]. For the sake of simplicity we will ignore the correction for the finite duration of the repetition period.

Since the delay \( t_1 \) is scanned over many orders of magnitude, it is convenient to plot the correlated charge \( \Delta Q \) as a function of \( \log_{10}(t_1/t_0) \) as in figure 1 (\( t_0 = 1 \) s). The QTS peak corresponding to \( \tau = 10^{-4} \) s has been simulated by a Gaussian

\[
y = B \exp \left[ -\frac{(x-x_c)^2}{2w^2} \right], \tag{4}
\]

after defining new variables \( x = \log_{10}(t_1), x_c = \log_{10}(t_{1m}) \), \( y = \Delta Q(x), w = \log_{10}(\Delta t) \) and \( B = 0.17Q_0 \). Such an approximation does not apply to the entire QTS peak, nevertheless the Gaussian width \( \Delta t \) of any QTS peak corresponding to a discrete energy \( E_{pi} \) is correctly expressed via \( w = w_0 = 0.335 \). Let us now consider a spectrum of energies \( E_{pi} \) or in terms of equation (2) a spectrum of time constants \( \tau_i(E_{pi}) \). In order to avoid numerical integration when expressing the signal \( \Delta Qs \) due to a continuous spectrum of time constants [11], we will instead consider a discrete spectrum of energies \( E_{pi}(i = 0, \ldots, n) \) spread around an average energy \( E_{pC} \). Then the task is to calculate the sum of individual contributions \( \Delta Q_i(\tau_i) \) to the total signal \( \Delta Qs \) in the \( x-y \) plane. Here only the case of a constant pre-exponential factor is treated, in general both \( \Theta_0 \) and \( E_{pC} \) may be distributed as a consequence of the disorder.

Given the values of the width \( w \) (variance) and \( x_c \) of the total spectrum, the simulation of \( \Delta Qs \) versus \( \log_{10}(t_1) \) starts with defining amplitudes \( \gamma_i \) of \( n + 1 \) components of a Gaussian distribution of discrete time constants (expressed as \( x_{mi} = \log_{10}(t_{1mi}) \)), falling in the interval from \( x_c - 2w \) to \( x_c + 2w \) and separated one from another by \( \Delta x = 4(x_c - w)/n \). Now the varying amplitudes \( \gamma_i \) of individual peaks are obtained by the help of equation (4) taking a constant value of \( B \). Finally the sum of individual QTS peaks \( \Delta Q(x_{mi}) \) (defined by equation (1)) is constructed:

\[
\Delta Q_s = \sum_{i=0}^{n} \Delta Q(x_{mi}). \tag{5}
\]

For assessing each QTS peak \( \Delta Q(x_{mi}) \) the delay \( t_1 \) was ‘scanned’ from \( 2 \times 10^{-6} \) s to 0.6 s. In figure 2 simulated spectra are shown (\( n = 10, 20, 40; \) \( w = 1.5; x_c = -3 \)), along with the values of the ‘observed’ widths \( w^* \), the error amounting less than 10% for the ‘density’ of levels \( n > 10 \). For completeness, the expected evolution of the total spectrum amplitude with variance \( w \) for a given value of \( n \) is illustrated by figure 3. A more detailed analysis of how to correct the error will be published elsewhere.
As a matter of fact we are interested in the variance of the energy distribution \( \Delta E_p \), rather than in the variance \( \omega \) of the distribution of the time constants. If dealing with a continuous distribution of time constants, then for each time constant \( \tau \) of the spectrum given by equation (2) one can write

\[
\tau \approx \log_{10}(\tau) = \log_{10}(|\tau|) + \log_{10}\left[ \exp\left( \frac{E_p}{kT} \right) \right].
\]

or \( x \approx x_0 + E_p/(2.3kT) \), after introducing the quantity \( x_0 = \ln(|\tau_0|)/2.3 \). This leads to the desired relation between energy \( E_p \) and \( x \):

\[
E_p \approx 2.3(x - x_0)kT.
\]

Evidently it is not possible to deduce the absolute value of any energy from a single scan of the time window \( t_1 \) at a constant temperature, as long as the quantity \( x_0 \) remains unknown. However, it is always justified to rewrite \( w \) as a difference of two quantities \( x_1 \) and \( x_2 \), respectively (\( x_1 > x_2 \)). Put in other words, for a given \( w \) one is able to transform the \( x \) axis to an \( E_p \) axis, thereby obtaining a valuable formula for the variance of the energy distribution:

\[
\Delta E_p \approx 2.3wTkT.
\]

If the most probable energy \( E_{pm} \) is demanded, one needs at least two isothermal QTS scans at two different temperatures \( T_1 \) and \( T_2(T_2 > T_1) \), with signals peaking at \( x_{1} \) and \( x_{2} \), respectively. Denoting the absolute value \( |x_{1} - x_{2}| \) by \( \delta x \), we obtain an expression for \( E_{pm} \) that reads

\[
E_{pm} \approx 3.8w(\frac{1}{T_1} - \frac{1}{T_2})^{-1}.
\]

In what follows we will present experimental data and exemplify how to use the analysis described above.

There is in principle the possibility of using thermal scan QTS, monitoring the temperatures of QTS maxima corresponding to different delays \( t_1 \approx \tau(T_{\text{max}}) \). However, the error connected with transforming the width \( w \) (variance in K) to \( \Delta E_p \) is now much higher, keeping in mind that for a given \( T_0 \) the variance \( w(T_0) \) of any thermal QTS peak due to a discrete energy \( E_p \) is proportional to the latter. This is the reason why a set of isothermal QTS scans is preferred, the option of finding the thermal activation energy via an Arrhenius’ plot as \( \ln(t_{\text{max}}) \) versus \( (kT) \) becomes evident after a simple manipulation of equation (9).

4. Results and discussion

A set of representative QTS spectra of a Ag/CuPc/ITO diode under different biases \( U \) of the ITO substrate obtained in response to both positive and negative pulses \( \Delta U \) are reproduced in figure 4. There is a faster process ‘A’ giving rise to peaks of comparable heights and opposite signs when reversing the polarity of the pulse. Evolution of the process ‘B’ is observed only for reduced negative biases and positive polarity of the pulse, conditions mediating injection of holes from the ITO anode to CuPc. Next we concentrate on the process ‘A,’ taking place close to equilibrium, i.e. under weak injection conditions. Apart from the comparable heights of the peak ‘A’ for opposite polarities of the pulse at a given reverse bias, two remarkable features of the peak are worth mentioning:

(i) for positive \( \Delta U \) the peak is positioned at a longer delay \( t_{\text{1.s}} \) compared to the case of \( \Delta U < 0 \),

(ii) the peak ‘A’ is much broader than that simulated for a discrete energy level (solid and dashed line), indicating the presence of a distribution of the relaxation time (energy). When starting the reconstruction of the distribution function, only the response belonging to negative \( \Delta U \) has been taken into account, thereby eliminating any interference with the process ‘B.’ As documented by figure 5, this set of QTS spectra matches fairly the case of the Gaussian distribution function, expressed via equation (4) with \( w \approx 0.81 \).

This finding served as the motivation for considering first the presence of true interface dipoles at the CuPc/ITO interface, in compliance with the model presented by Baldo and Forrest [8]. Our attempt to modify their qualitative energy band diagram resides in admitting at least a partial rotation.
of originally misaligned permanent dipoles located in the thin film of thickness \( L \) after applying an external electric field \( F \).

In addition, now the dipole strength of these ‘interface’ dipoles is supposed to be comparable to that of immobile permanent dipoles of individual molecules in the bulk of CuPc. Consulting the monograph by Fröhlich [12], the amount of polarization (density of dipole moment) is given by

\[
P_0 = \frac{p^2 N_0}{3kT} F,
\]

where \( N_0 \) stands for the spatially uniform density of dipoles. Before a further treatment of the variance \( w \), let us estimate the concentration of molecules, or bulk dipoles in CuPc. Considering the molecular structure \( \text{C}_{32}\text{H}_{16}\text{N}_{2}\text{Cu}_4 \), we obtain a molecular weight \( m^* = 575 \text{ g} \). Then for the density \( \rho^* = 1.65 \text{ g cm}^{-3} \) the corresponding molar volume is \( V_m = m^*/\rho^* = 348 \text{ cm}^3 \). In order to obtain the density of molecules \( N_{\text{CuPc}} \), we simply consider the ratio \( N_{\text{CuPc}} = N_A/V_m \). Here \( N_A = 6.02 \times 10^{23} \text{ mol}^{-1} \) is Avogadro’s number, the density of molecular dipoles amounting to \( N_d = N_{\text{CuPc}} \approx 1.7 \times 10^{23} \text{ cm}^{-3} \).

In order to relate this result with the experimentally detected charge, we take \( \varepsilon = 2.5, \Delta Q_{\text{em}} \approx 8 \times 10^{-13} \text{ C}, F = \Delta F = \Delta U/L = 3 \times 10^5 \text{ V m}^{-1}, \rho = 1.43 \text{ Debye}, \) and assume freely rotating molecular dipoles of CuPc. The \( \varepsilon \) and \( \rho \) values assigned to a CuPc monomer have been taken from the paper by Sakakibara et al. [13]. Applying equation (10) with \( P_0 = \Delta Q_{\text{em}}/(0.17 \text{A}) = 2.21 \times 10^{-5} \text{ C m}^{-2} \), the calculation yields \( N_d = 4 \times 10^{21} \text{ cm}^{-3} \). This dipole density corresponds to a single energy \( E_p \), ignoring the presence of the spectrum of energies. Nevertheless, it is significantly higher than that of CuPc molecules in a perfect crystal, moreover, the density of the CuPc film is definitely lower than that of the crystal (voids). Under the assumption of the same constant double moment on all the molecules, we have abandoned further attempts to treat the charge relaxation (process ‘A’) in terms of orientation of intrinsic dipoles uniformly distributed throughout the layer.

Seeking for a more adequate approach, we inspect consequences of slow hopping of charge carriers within a Gaussian DOS in a disordered organic solid. As stated by Jonscher [14] in his review article on dielectric relaxation in solids, the hopping transport leads to a dielectric relaxation of the ‘charge carrier’ type rather than of the ‘dipolar’ type. If this is the case, then there is a dielectric loss monotonously changing with time or frequency of observation, without showing any maximum. However, there is abundant evidence for peaks of dielectric losses in disordered solids with the hopping (small polaron) transport provided there is a metallic component with alternating valence (V, Fe, Cu). To exemplify this type of dielectric loss (in a two-level system), we quote the published work on ac relaxation in cuprate glasses [15] and strongly correlated \( \text{Gd}_2\text{Sr}_{1-x}\text{FeO}_3 \) oxides.16 However, the QTS features of the process ‘A’ mentioned above under items (i) and (ii), respectively, contradict the main signature of the dielectric relaxation, which is the equality of the time constants of the response for opposite polarities of the excitation pulse.

Coming back to the dominant charge relaxation in CuPc under reverse biases, we try to view the variance \( w \) of the Gaussian distribution from the QTS data as reflecting the variance \( \sigma_d \) (in eV) of a DOS generated by guest dipoles not undergoing reordering (reorientation) after applying a bias

\[
\sigma_d(a, p, \varepsilon, c) = \frac{3.06}{a^2 \varepsilon^{2/3}} p.
\]

Before calculating the relative concentration \( c \) we need to transform \( w \approx 0.81 \) (from figure 5) to \( \sigma_d \) by virtue of equation (8) with \( \Delta E_p \) replaced by \( \sigma_d \). In this way (tolerating \( \approx 10\% \) error) one obtains \( \sigma_d \approx 0.048 \text{ eV} \pm 0.005 \text{ eV} \) so that after choosing \( a = 3.35 \text{ Å} \) and the theoretical value \( p = 1.43 \text{ Debye} \) (both from [13]) one obtains \( c \approx 0.17 \). It is interesting to compare \( \sigma_d \approx 0.048 \text{ eV} \) as obtained for the CuPc thin film from the QTS data with the one predicted in figure 3(b) of [9] for \( p = 1.43 \text{ Debye} \) (cf figure 6). The dipolar property of our CuPc thin films resembles strongly that of molecular glasses TAPC, TPD, TTB [9]. If our reasoning is correct, then the volume fraction of polar CuPc molecules may correspond to about 17% of the total number of molecules in the CuPc thin film. A comprehensive review of the effects of polar molecules and dipolar traps on the transport of charge in molecular materials has been published by Sworakowski [17].

The situation is complicated by the possible presence of both \( \alpha \) and \( \beta \) polymorphs in the CuPc films. Our preliminary \textit{ex situ} results on x-ray diffraction are indicative of either the prevailing \( \alpha \) (crystalline) modification of CuPc or of a heavily disordered \( \beta \) phase. More attention should have been paid to \textit{in situ} evaluation of the morphology of the films prior to Ag deposition.

Considering again the finding envisaged by figure 4, namely the relaxation speed after positive excitation pulses at a given bias is lower than that in response to pulses of negative polarity. This behaviour is consistent with the dynamics of filling and emptying traps while using pulse excitation of a hole-transporting semiconductor [18]. After a positive pulse there is emission of holes from the localized states, a process which is slower than the capture of holes to these states after having emptied the states by the increased negative (reverse) total bias during a negative pulse.
5. Conclusions

In conclusion, we have derived a simple formalism for analysing QTS coming from charge relaxation processes characterized by a Gaussian distribution of relaxation times, with emphasis put on distributions in thermal activation energy rather than in the pre-exponential factor. Exploring QTS response of single-layer Ag/CuPc/ITO diodes, an evidence for a Gaussian distribution of time constants assigned tentatively to dipoles (polar molecules) in CuPc has been provided. The theory presented by Dieckmann et al [9], based on the dualism of (probably immobilized) dipoles and related DOS, has been utilized in order to find the variance of the DOS distribution \( \sigma_d \approx 0.048 \) eV from the width \( w \approx 0.81 \) of the measured QTS signal redrawn as a \( \Delta Q \) versus \( \log_{10}(t_1) \) plot. In addition, after taking the involved dipole moment as equal to that of a CuPc monomer, the relative concentration of dipoles (polar molecules) has been estimated as \( c \approx 0.17 \). This may indicate a severe compositional disorder (phase separation) in the CuPc thin films. Whatever the case, the anticipated dipolar property of the CuPc film resembles strongly that of molecular glasses. One should even be capable of determining the energy where the hopping transport takes place after conducting a series of QTS measurements at different temperatures. Since one cannot exclude effects of traps of other than dipolar origin, the thermal activation energy is to be correlated with \( \sigma_d \) within the framework of the model suggested by Bässler [19].

References