

Monte Carlo simulation of charge-trapped effects on dispersive electronic transient transport

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By means of the Monte Carlo technique the possible mechanism responsible for charge trapping at the region of photogeneration of nonequilibrium charge, in dispersive transient transport measurements, was studied. The typical curves for dispersive transport with the effect of trapped charge were simulated. The temperature dependence of the simulated currents and the corresponding experimentally obtained currents, measured by the time of flight technique, were compared. A good agreement between experimental and simulated results was obtained, and a model for the mechanism of charge trapping at the photogeneration region is proposed. [S0163-1829(98)03045-8]

I. INTRODUCTION

A. Measurement of dispersive transport

The time-of-flight (TOF) technique is a very useful experimental tool for the study of transport properties in solids, such as drift mobility, density of traps, activation energy, etc. It is important in the study of disordered solids because low mobility and high resistivity are an obstacle for the application of steady-state techniques, such as the Hall-effect technique. In the last two decades, a great amount of work has been done on transport in disordered solids, such as α -As₂Se₃,¹⁻³ α -Se,¹⁻³ α -Si:H,⁴ by using the TOF technique.

In an experiment of TOF, a pulse of strongly absorbed light, incident near an electrode, generates nonequilibrium electron-hole pairs. Depending on the polarity of the constant applied bias voltage, holes (electrons) are absorbed at the nearest contact leaving a sheet of electrons (holes) to move to the far electrode. Changes of the field across the sample, induced by the moving packet of charge, generates a current as the voltage source attempts to maintain a constant voltage.

Electrical transport in disordered solids is dispersive, and it has been demonstrated by several authors,⁵⁻⁹ that the transient current obtained by the TOF technique can be described as

$$I(t) = \begin{cases} t^{-(1-\alpha_1)}; & t < t_\tau \\ t^{-(1+\alpha_2)}; & t > t_\tau. \end{cases} \quad (1)$$

α_1 and α_2 are the dispersion parameters at short times and at long times, respectively. By plotting the current in a log-log scale, the transit time is determined as the crossing point between two straight lines. The transient current behavior is understood as being due to the packet of electrons (holes) becoming spread out or dispersed to a much greater extent than one would expect from diffusion alone.⁵ Therefore, it is referred to as dispersive (non-Gaussian) transport.

B. The models

Montroll and Scher 1975,⁵ proposed a transport model that explained the dispersive features of the transient currents in amorphous solids. They used a simple hopping model: the continuous time random walk.^{5,6} After that, some authors proposed other models such as the trap-controlled hopping model,⁷ and the multiple trapping model.^{8,9}

When transport is by simple hopping, the charge carriers move between localized states of the band tails by tunneling,¹⁰ and the random distribution of the hopping distances gives rise to the dispersion of the transit time. However, in order to explain the existence of a high activation energy (deep traps) it is necessary to include the presence of deep traps randomly distributed in space. The temperature dependence of the transit time is due to the activation energy from the trapping center, and the dispersion of the transit time arises from the random distribution of hopping distances. In both cases, simple hopping and trap-controlled hopping, it seems that the dispersion parameters α_1 and α_2 are temperature independent.

When temperature dependence of the dispersion parameters is present, an explanation in terms of the hopping models is obtained by adding a distribution of activation energies to the distribution of distances between hopping sites.

A different conception on the origin of the dispersion of transit time is given by the multiple trapping model, regarding the distribution of activation energies as the origin of the dispersion of the transit time. In this model, charge carriers move by repeatedly trapping from extended states to localized states, and releasing from the localized states to extended states. The dispersion of the transit time is caused by the energy distribution of localized states, or activation energies. It has been demonstrated that the multiple trapping transport model and the continuous time random-walk model are equivalents.^{1,8,11}

From the above models, one can say that it is apparent that temperature dependence of the dispersion parameters α_1 and α_2 is due only to the activation energy distribution (trap distribution). It means that if the disorder in the energy separation is negligible, the α_1 and α_2 parameters can be weakly dependent on or even independent of the temperature.¹²

C. Trapped charge effects

However, the determination of a transit time from the transient current curves is possible only when well-defined transient currents are obtained. In some cases the behavior of the transient current is not described by Eq. (1), but it presents a maximum that avoids the determination of the transit time, even when transport remains dispersive. This phenomenon has been studied by Abkowitz and Scher¹³ in the frame of the continuous time random-walk model for dispersive transport. They explained the presence of a peak of current as caused by the trapping of charge in the region where non-equilibrium charge carriers are photogenerated. Consequences of this trapping of charge results in an effective time delay of carriers, and it was found that the effect of trapping and releasing the charge carriers in that region can be well described by introducing a time-release distribution function^{13,14}

$$\phi(t) = \frac{(t_1 + t_2)}{t_2^2} (1 - e^{-t/t_1})e^{-t/t_2}. \quad (2)$$

Here t_1 and t_2 are the parameters controlling the raising and decreasing of the distribution function $\phi(t)$, respectively. Equation (2) represents the normalized probability for a carrier to reach the transport states (sample) at time t after being trapped at the photogeneration region.

In the present work we simulate the dispersive transport by using a model, proposed by Murayama,¹⁵ based on the idea that the electronic transient transport can be treated as a random walk of particles on percolation clusters, as we will see in next section. In order to get the effect of the trapping and releasing of charge, we adapted the distribution function given by Eq. (2) to the Murayama model. This minor modification allows us to compute the time delay of the nonequilibrium charge carriers before they enter the ‘‘bulk,’’ where the transient current is induced.

On the other hand, a relation between the t_1 and t_2 parameters does not exist that indicates the temperature dependence of the peak of $\phi(t)$. Simulating the shape of this function with the Murayama’s model we obtained a good agreement with the experimental results observed when the temperature changes.

II. THEORY

A. The transport model

Dispersive transport has been studied on the frame of the theory of stochastic processes, where it does not depend in a detailed way on a conduction mechanism. In this case, charge carriers are considered as subject to a time event distribution, events which can be hopping of a particle from one localized state to another, trapping from a conduction state to a deep state (trap), activation from a deep state to the con-

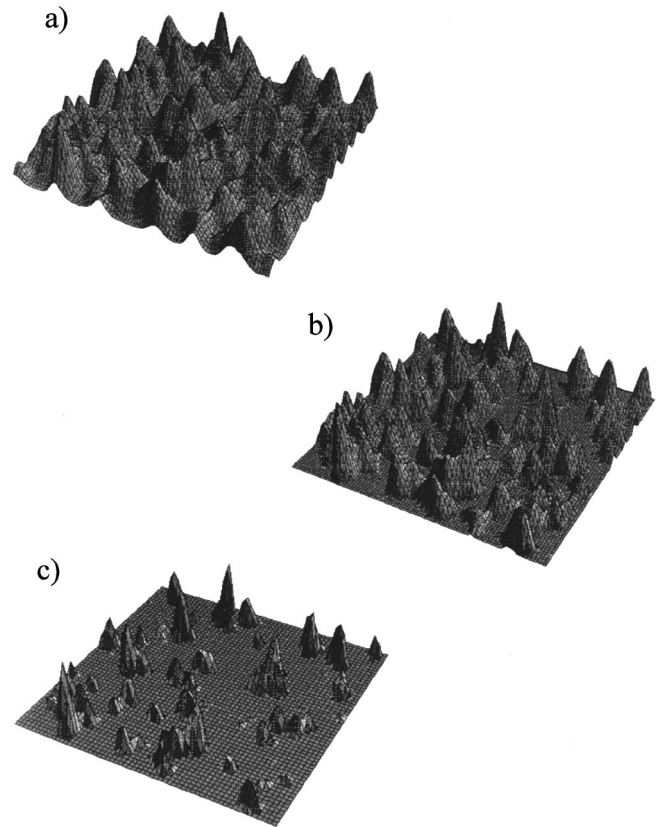


FIG. 1. Random surface with different energy levels (a), (b), and (c). The flat regions at the bottom correspond to the constant energy level.

duction states, etc.^{12,14} The movement of charge is via a sequence of time events in the presence of an applied electric field. Due to disorder, the event time can be a random variable, characterized by the probability $p(t)dt$ that the time for an individual event is between t and $t + dt$. The accumulated sequence of these events in the motion of a charge carrier can be viewed as a continuous time random walk.¹²

Thus, the main tools for the analysis of dispersive transport are the time event distribution and the disordered arrangement of sites or energy distribution. In this work we use a model for dispersive transport which is based on percolation theory and the multiple barrier transport mechanism.¹⁵ This model fits very well the nonlinear temperature dependence of the dispersion parameters, α_1 and α_2 for a -Si:H at low temperatures.¹⁶ The basic assumption of this model is that fluctuations of the band edge in an amorphous system can be closely related to a distribution of barrier heights between hopping sites.^{1,6,17–22} We illustrate a three-dimensional frame, two-dimensional in space and one-dimensional in energy, to show the space-energy fluctuation of the conduction band edge in Fig. 1. Flat regions determine the limited space where an electron can move for a given energy. As can be seen in Fig. 1(a) electrons are constrained to move inside small regions of space at low energies (temperature) and by increasing energy these regions eventually connect one another. When the flat regions are connected such that an infinite path is created, the percolation threshold is reached, then electrons can move over the entire system [Fig. 1(c)].

In that sense, percolation theory is a useful tool for the study of disordered systems, and it predicts that in a random lattice of sites a percolation path will be formed if at least a fraction p_c of the total number of sites is connected (occupied). A percolation path is a cluster of connected sites that span all the system.

By analogy between percolation by sites and an amorphous solid, a random distribution of localized states, the existence of an infinite cluster formed by connected states is determined by the fraction of occupied sites (p). The density of states for an amorphous solid is generally accepted as an exponentially dependent function of energy, so that an analytic expression for the fraction of occupied sites can be¹⁶

$$p = \int_{-\infty}^E g(E) dE \Big/ \int_{-\infty}^{E_M} g(E) dE = e^{(E-E_M)/kT_c}, \quad (3)$$

where E_M is the mobility edge, T_c is a slope of the density of states.

The barrier heights and the hopping distance distributions can be substituted by the topological disorder of a percolation cluster formed by the localized sites of the system. Also the movement of a particle by thermally activated hopping over multiple barriers can be substituted by the random walk of a particle over a disordered arrangement of sites.^{5,16}

The influence of spatial disorder, experimental conditions of temperature and applied voltage, over the nonequilibrium charge packet is considered by the jump probability for random walk. Murayama¹⁶ used the jump probabilities

$$P_{\pm x} = e^{\pm eFa/2kT} / (4 + e^{+eFa/2kT} + e^{-eFa/2kT}), \quad (4)$$

$$P_{\pm y} = P_{\pm z} = 1 / (4 + e^{+eFa/2kT} + e^{-eFa/2kT}),$$

for a particle in a cubic lattice, with a biased applied electric field (F) in the x direction.

Simulating the transient currents for different applied bias electric fields, cluster size, and fraction of occupied sites, it can be shown that for dispersive transport on percolation clusters, the fraction of occupied sites is proportional to temperature¹⁶

$$p = sT/T_c. \quad (5)$$

In order to include the effects of the traps at the photogeneration region, we modified Murayama's model by including a distribution of the trapping probability for the carriers before they reach the percolation cluster. It means that the charge carriers now have to travel through a region of traps where they can be trapped, and released after some time, according to a distribution of the probability given by Eq. (2).

The photogeneration region corresponds to the interface sample contact when the sample is sandwiched by contacts, one of these transparent to the incident light.^{13,14} Also, it is possible to have two contacts over the same face and illuminate a thin region near a contact.²³ In both cases the nonequilibrium charge carriers are generated near a contact and dragged to the opposite side, which is considered in the used model to simulate the transient transport.

B. Simulation of transient currents

The process of simulation of dispersive transport is the same used by Murayama. First, one has to construct the percolation cluster (connected sites) formed in a random distribution of localized energy levels for an amorphous system, in which case a cubic lattice is used for simplicity. We used the Hoshen-Kopelman algorithm to construct the percolation clusters.²⁴ Percolation paths are primarily formed in order to have a cluster where the carriers can move by random walk. Charge carriers (particles) are introduced into the percolation cluster from a face of the prism shape sample and are collected at the opposite face, simulating the electrode configuration in the TOF technique. In order to include the charge-trapping effect, we consider that each particle is trapped before it enters into the percolation cluster, so that the time it remains trapped is calculated from the distribution of probability $\phi(t)$. Once the releasing time (t_r) is determined, the particle is introduced into the percolation cluster and then it starts moving in a random walk at time t_r . A particle can move only inside the percolation cluster, i.e., if the jump probability is such that a particle must jump to a site that does not belong to the percolation cluster, then the particle does not move, and the number of step increments and the jump probability is recalculated. After n steps the mean position of the charge packet, the number of particles inside the cluster $N(n)$, and the mean velocity $\langle V(n) \rangle$ of the charge packet are calculated. This process is repeated for different values of steps until the total number of particles inside the cluster is zero ($N(n)=0$), i.e., when all the particles have reached the opposite side of the cluster (absorbing boundary). The induced current after n steps is calculated from the relation $I(n) \sim \langle N(n) \rangle \langle V(n) \rangle$, as in Ref. 16, and averaged for different forms of percolation cluster (topology) with the same fraction of occupied sites p .

On the other hand, for the trapping-releasing events at the interface, a crossing time distribution $\Phi(n)$ is defined. This distribution is constructed by calculating the number of steps (time) needed for each particle to cross an entire percolation cluster and then counting the number of particles that have reached the opposite side of the cluster after n steps. The normalized distribution $\Phi(n)$ can be interpreted as the probability for a particle to cross the percolation cluster after n steps.

III. RESULTS AND DISCUSSION

Figure 2(a) shows a plot for the simulated transient current $I(n)$, and the number of particles moving inside the cluster $N(n)$, as a function of the step number (n). Transient current curves can be fitted by the expression given in Eq. (1). As can be seen, the main features of the transient current are reproduced by Murayama's model for dispersive transport. The number of particles $N(n)$ inside the cluster at time $0 \leq t \leq t_r$ is constant and equal to the total number of particles, and it diminishes each time a particle arrives to the opposite side of the percolation cluster. The transient current decreases as $t^{-(1-\alpha_1)}$ for $t < t_r$, and has a faster decaying as $t^{-(1+\alpha_2)}$ for times greater than t_r , caused by the diminishing of the number of particles contributing to the induced current.

When the effect of the charge trapped at the photogenera-

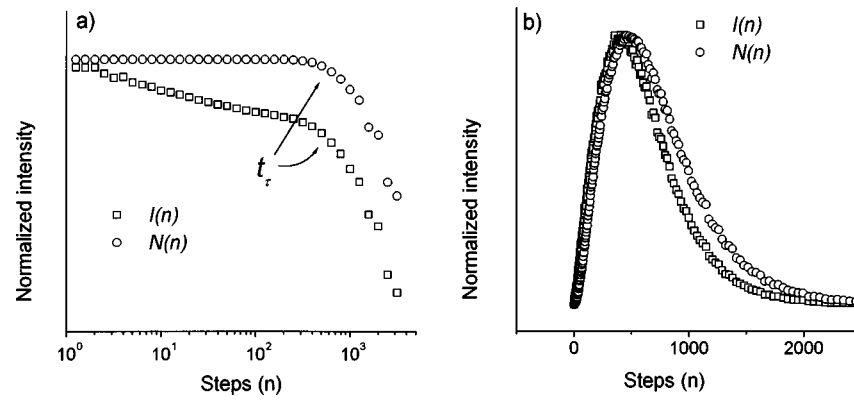


FIG. 2. Number of particles inside the cluster, and transient current, after n steps, for the cases (a) when no charge is trapped at the photogeneration region, and (b) when all charge is trapped at $t=0$.

tion region is present, as in Fig. 2(b), the number of particles at time $t=0$ is zero, i.e., at that time all particles are trapped at the photogeneration region.¹⁴ As time evolves the probability for a particle to reach a transport state (infinite cluster) augments, and consequently the number of particles inside the percolation cluster increases with time. In Fig. 2(b), it can be seen how the transient current depends on the number of particles inside the cluster, when all particles are inside the cluster the induced current is maximum, and as particles reaches the absorbing boundary (contact) the current decreases. As seen, transient current is highly dependent on the time a particle delays to enter into the cluster. To clarify this effect, we simulated the case when a fraction of the total number of particles, forming the packet of the charge, goes into the cluster without being trapped, and the rest of the particles are trapped. It means that the time-release distribution function injects a fraction n_0 of particles a time $t=0$ and a fraction $(n_{\text{tot}} - n_0)$ of particles at $t > 0$, with a time distribution given by $\phi(t)$ [see Fig. 3(a)]. In Fig. 3(b) it is appreciable how the transient current changes with n_0 . In the limit case when $n_0=1$, no particles are trapped at $t=0$, the transient current has a characteristic shape of dispersive transport, as seen in Fig. 2(a) and the inset of Fig. 3(b); i.e., it is always decreasing and described by Eq. (1). For the opposite limit, when $n_0=0$ and all particles are trapped at $t=0$, the transient current starts increasing until it reaches a maximum value, and then it decreases when particles come out of the cluster.

Thus it is clear that the peak of current is due to the presence of trapped charge at the photogeneration region, which is released with a time delay given by $\phi(t)$. Moreover, the shape of the simulated current is due to this characteristic time-release distribution of particles. It can be shown by comparing the shape of $\phi(n)$ with the corresponding simulated transient current $I(n)$, as illustrated in Figs. 4(a) and 4(b). The inset in Fig. 4 shows the peak position values, for both $\phi(n)$ and $I(n)$, as a function of the t_2/t_1 rate. From these curves, it is evident that the shape of the transient current, and its peak position, are due to the time-release distribution. Furthermore, it was observed that by increasing the rate t_2/t_1 , the simulated current peak position shifts to larger values at the same time that the corresponding intensity of the simulated current $I(t_m)$ decreases [see Figs. 4(a) and 4(b)]. This behavior on the transient current is similar to that observed experimentally by other authors in chalcogenide multilayers²⁵ and polycrystalline CdTe thin films²³ measured by the TOF technique at different temperatures (see Fig. 5). The shifting of the peak position t_m for the experimental case is similar to that of the simulated currents. Also, in both cases, experimental and simulated, the intensity of transient currents at time t_m decreases with temperature (p), as seen in Figs. 4(b) and 5. This means that the rate t_2/t_1 acts as a temperature parameter and that contribution from the trapped charge is smaller when temperature is lowered. In other words, the nonequilibrium charge created in the isolated clusters has less probability to enter into the transport

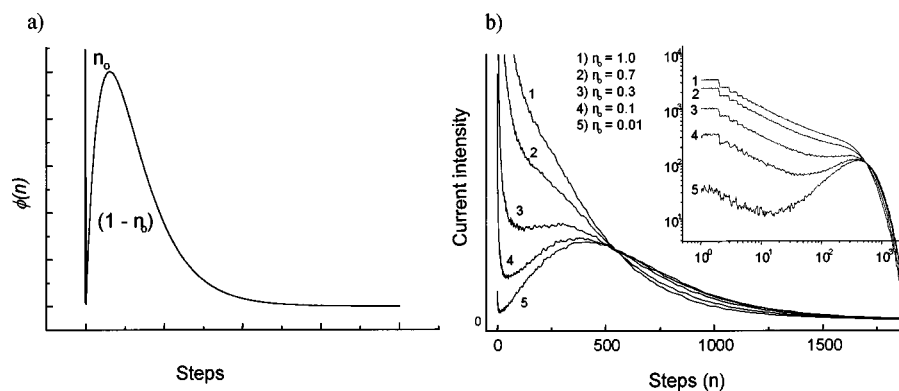


FIG. 3. (a) Time-release distribution function including the directly injected fraction of charge (n_0) and the trapped fraction of charge $(1 - n_0)$. (b) Simulated transient current for different values of n_0 .

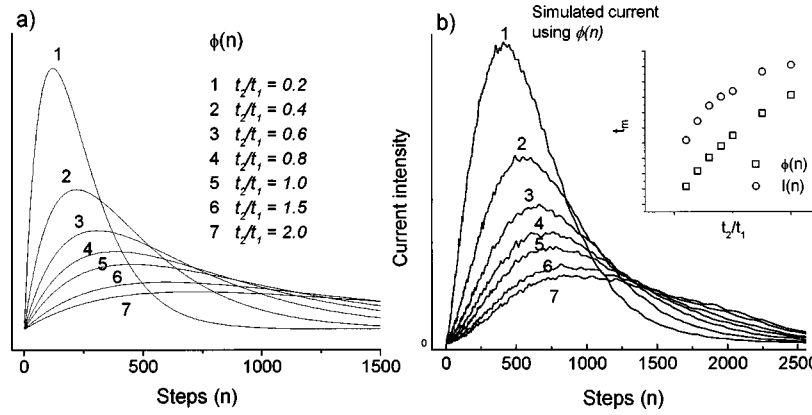


FIG. 4. (a) Variation of the time-release distribution function $\phi(n)$ for different values of the parameter t_2/t_1 , and (b) the correspondent simulated current $I(n)$. (c) Peak position as a function of t_2/t_1 for both $\phi(n)$ and $I(n)$.

states because the energy supplied (temperature) is smaller. However, we cannot obtain an analytical expression for t_2/t_1 as a function of temperature to study the t_m shifting for the experimental case.

Thus, it is necessary to find a time-release distribution function for the nonequilibrium charge carriers with an explicit temperature dependence. In order to do that, we realized that the region where the charge is trapped consists of localized sites randomly distributed, which can be deep traps and shallow states. Then one expects that the nonequilibrium charge carriers must travel across a random distribution of localized states and eventually can be trapped in a deep trap. One can also suppose that particles move by random walk into a cluster of localized sites, in the same way that they move on the transport states of the sample.

Thus, there exists two regions where the particles can move and induce a transient current: first they have to cross a very thin space, the photogeneration region, of distance $\Delta L \ll L$, and then they travel through the conduction states of a sample of thickness L . Because both regions have different time scales for the trapping-releasing process, the cumulative effect of the crossing of particles from one region to the other is well described by the time-release distribution $\phi(t)$ associated with the charge injection from the interface.

A more clear visualization of this mechanism is illustrated in Fig. 6.²⁶ Suppose that we have a region A, which has low

mobility, and a region B with a high mobility. If we introduce a packet of nonequilibrium charge carriers, at time $t = 0$, subject to an external applied bias electric field, this charge packet moves slowly in the first region ($t = t_1$) and eventually it will reach the A-B interface at time $t = t_2$. When the charge packet moves in region A, this movement induces a current with the characteristics of dispersive transport. As the charge arrives to the second region B, the current must increase because this region has a higher mobility. Thus the form of the current induced by the charge moving through the interface A-B will be an increasing function of time, and consequently the current induced by the movement of charge through the region B will not be described by Eq. (1), even if transport remains dispersive. In this case the current is influenced by the delayed injection of charge, from region A to region B, which gives to the current the charac-

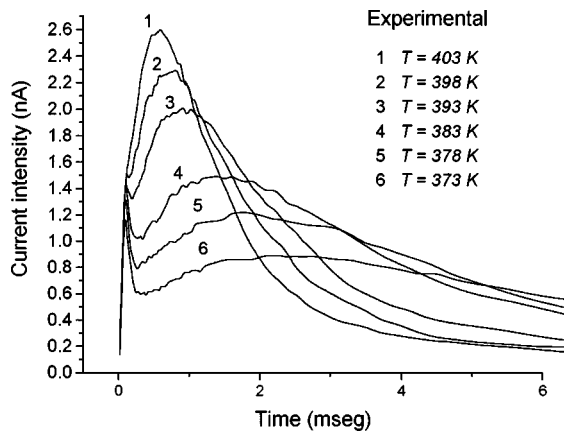


FIG. 5. Transitory currents, experimentally measured, for polycrystalline CdTe thin films at different temperatures (Ref. 23).

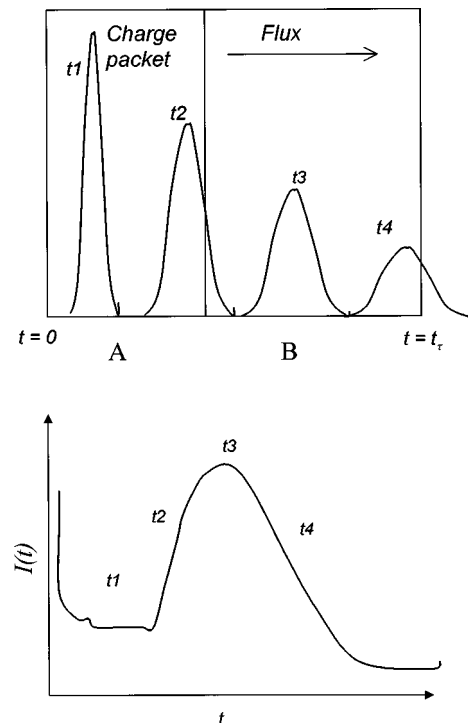


FIG. 6. Schematic representation of the transient current induced by a packet of charge moving through layers of different mobility.

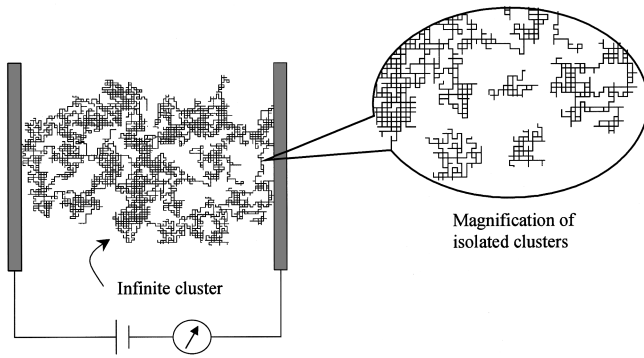


FIG. 7. Schematic representation of the infinite cluster of connected states in a sample, with the isolated clusters magnified.

teristic shape shown in Figs. 5 and 4(b).

Thus, it means that the experimental set of a sample in an experiment of TOF must be considered as composed by a region of low mobility (injection region), a high mobility region (sample), and an absorbing region (absorbing electrode). The effect of charge trapping at the photogeneration region is associated with the delay time of a charge crossing through a low mobility region.

From the point of view of the present model, the low mobility region, or trapping region, can be associated with the presence of localized states forming finite clusters not connected to the percolation cluster or percolation path (see Fig. 7). However, we think that even the isolated clusters are separated from the main cluster by a long hopping distance or a large energy difference, it is possible for the nonequilibrium charge carriers to jump out of this region by waiting more time. It means that, if we photogenerate nonequilibrium charge carriers, not all of them can reach the transport states at $t \cong 0$, but they can be released at longer times, resulting in a transient current similar to that represented in Fig. 3(b) for $n_0 \neq 0$. This mechanism is more probable in polycrystalline systems because of the random distribution of barriers created by the presence of charge trapped at the grain-boundary surface levels.

Also, the time delay of charge carriers at the photogeneration region is important when transient photocurrents are studied in multiple layered systems. In Ref. 26 it is shown how the transport described by Fig. 6 is reproducible for

organic-organic interfaces which have different mobility values.

Returning to the simulation of dispersive transport, we have seen that the shape of the simulated transient current is due to the time-release distribution for the trapped charge at the interface $\phi(t)$. Also, the temperature dependence of $I(n)$ and $\phi(n)$ are similar, so we can study the behavior of the peak position of $I(n)$ for different temperatures by simulating the time-release distribution of a carrier crossing a percolation cluster for different values of p .

We used the same method of construction of percolation clusters²⁴ and calculated the time delay for each particle crossing the entire percolation cluster, resulting in a time-release distribution $\Phi(t)$. Figure 8(a) illustrates how $\Phi(t)$ has a similar behavior than that for the time-release distribution $\phi(t)$, as expected.

Simulation of different $\Phi(n)$ distributions was carried out, and the peak position for each curve was determined. Figure 8(b) shows the peak position values t_m , as a function of temperature, for the experimental and simulated case. Experiments were carried out on polycrystalline CdTe films by the TOF technique. It clearly illustrates an exponential energy dependence, which resembles an activation time of the form

$$t_m = \nu_0^{-1} e^{E_{\text{act}}/kT}. \quad (6)$$

E_{act} is assumed to characterize the energy depth for the traps at the interface sample contact.

IV. CONCLUSIONS

It was possible to simulate the charge-trapping mechanism in the region of photogeneration of nonequilibrium charge carriers, and its effect on the transient currents experimentally measured by the time-of-flight technique. The mechanism responsible for charge trapping can be explained as a consequence of the dispersion of charge carriers crossing through a low mobility region (photogeneration region) before they reach the conduction states of the sample. This low mobility region can be due to the presence of isolated clusters, formed by localized states, which act as traps when charge carriers are delayed. The photogenerated free carriers have to walk randomly into the space where they are created

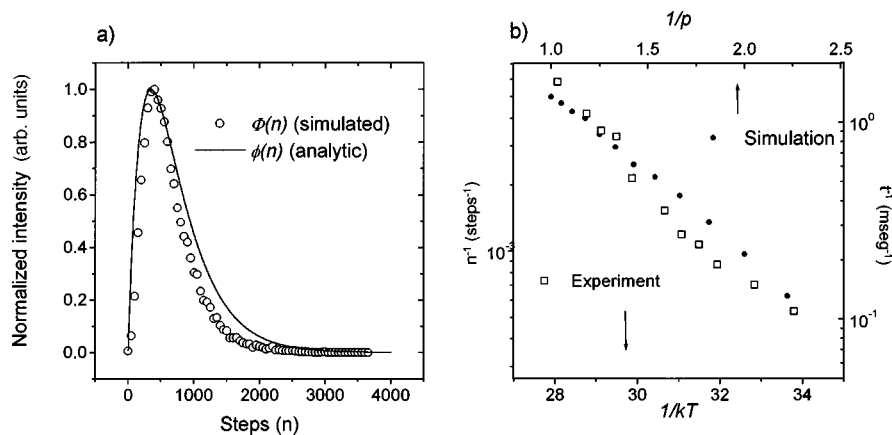


FIG. 8. (a) Comparison between the analytical time-release distribution $\phi(n)$ and simulated time-release distribution $\Phi(t)$. (b) Temperature dependence (p) of the peak position (t_m) for both simulated and experimental transient currents.

in order to reach the transport states, where they can move faster. Thus, the difference in time scale for events occurring in both regions (photogeneration and bulk sample regions) allows us to consider the charge packet arriving from the photogeneration region, to the bulk sample as a time-dependent injection function of nonequilibrium charge carriers. Furthermore, the time-scale invariance characteristic of dispersive transport mechanism, allows us to propose the application of this model to the study of phototransient currents in amorphous multilayers. Also, this work illustrates some ideas for the explanation of dispersive transport in polycrystalline semiconductors, as those of Fig. 5.

Also, we simulated the time-dependent injection of charge by forcing the nonequilibrium particles to travel through a percolation cluster before they reach the transport states. The same effects of a time-release distribution were obtained. From these results the current peak shifting with temperature was simulated, obtaining a good agreement with the experimental measurements.

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