Transient transport in disordered multilayers

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(Received 15 May 2000; published 4 June 2001)

In this work the main characteristics of transient photocurrents on disordered multilayers, as obtained by the time-of-flight technique, are simulated and compared to experimental data from the literature. The origin of the double-peak shape adopted by the photocurrents is discussed in this work, and it is shown that each peak is directly related to the time delay of charge carriers at the layer-layer and contact-layer interfaces, respectively. Two mechanisms of conduction are involved in the transport of carriers through the multilayer structure.

DOI: 10.1103/PhysRevB.64.014201 PACS number(s): 73.50.Gr, 72.10.−d

INTRODUCTION

In the last few years a renovated interest in the study of nonequilibrium charge transfer between disordered media has been recovered, being the cause of the great importance that a good photosensible material may have in the optoelectronics industry. The most interesting properties of these materials are the spectral sensitivity for charge carrier photogeneration, drift mobility, and the charge injection rate at interfaces. Recently, the deposition of amorphous multilayer structures has been done in order to improve some of these transport properties and to build devices with specific characteristics.[1] The transport properties of amorphous and disordered materials have been characterized by the canonical technique time of flight (TOF).[1,2] In this technique the sample is placed between two electrodes at constant bias, and then a pulse of strongly absorbed light near an electrode generates a sheet of electrons (or holes) which move to the far electrode. Changes of the field across the sample induced by the moving packet of charge generate a transient current as the voltage source maintains a constant voltage between electrodes. For dispersive transport in a single layer, the induced current has two rates of decay,[2,3] and in general it can be described by the following expression:

$$i(t) \approx \begin{cases} t^{-(1-\alpha_1)}, & t > t_\tau, \\ t^{-(1+\alpha_2)}, & t < t_\tau, \end{cases} \tag{1}$$

where α_1 and α_2 are the dispersion parameters at short and long times, respectively. The time needed for the carriers to cross the sample is known as the transit time t_τ.[2,3]

Since the former studies in As_2Se_3 for photocopy applications[4] in the 1970s to the most recent studies of organic-organic layers[1,5] and inorganic multilayers[6,7] in the early 1990s, the TOF technique has been of great importance in the study of the transport properties of materials. The same happens to the accepted theoretical models for dispersive transport in disordered systems where the universal models seems to be hopping (CTRW) (Ref. 3) and multiple trapping.[8] However, these models have been formulated with complicated mathematics for single layers only, and a generalization to include those systems composed with more than two layers is even more difficult.

Attempts to study charge effects at interfaces by generalized transport model based on CTRW were carried out by Abkowitz and Scher in 1977 (Ref. 9) by considering the interface contact sample as a time-dependent source of charge carriers. Assuming that the carriers in this region do not incorporate immediately into the transport states, they proposed a distribution function for the probability per unit time that a carrier at the interface incorporates into the transport states. However, the explanation of the time delay probability of charge at interfaces was given *a priori* with a function depending on two parameters controlling the rise and decrease of the probability function.[10] The time delay of charge carriers at interfaces of multilayers structures has been observed experimentally.[5,13,14] There are two possible reasons for the time delay of carriers at interfaces: first, the existence of energy barriers between interfaces due to either a mismatch in the mobility edge of amorphous layers or a different ionization potential in organic layers and, second, the presence of deep traps at the interfaces. In the first case, carriers need some time to tunnel through or to be thermally activated over the barrier. In the second one, the delay time is the time that carriers remain in the traps before they are thermally released. For the case of traps with a single energy level, the release rate is exponential with a characteristic time, the detrapping time,[2] which has an exponential dependence on the energy depth of the trap. The thermal activation of carriers over an interface barrier has also an exponential rate with a characteristic time, which depends exponentially on the barrier height. Recent efforts for the explanation of charge transport in single- and double-layer systems[10,11] and polycrystalline systems[12] have been done by several workers using different approaches. However, theoretical models that explain the main characteristics of the diffusion of nonequilibrium charge carriers in complex systems such as disordered multilayers are still lacking.

In this work we are interested on the off-equilibrium charge transfer between disordered layers of multilayer structures and especially how the time delay of carriers at interfaces affects on the general shape of transient photocurrents. Our intention is to provide good insight into this prob-

lem by simulating some properties of transient photocurrents that could be of interest for a future general dispersive transient transport model in multilayer systems. Also, in order to elucidate if our ideas are correct, we bring to the scene some experimental results of transient photocurrents for $Se/Se_{1-x}Te_x$ (Ref. 13) and a-Si:H/a-SiN_x:H (Refs. 7 and 14) multilayers. In these works the measured photocurrents show a double-peak shape, which remains as a general characteristic for these systems and is explained in terms of the existence of two mechanisms of transport that may occur at the same time. One mechanism is hopping through localized states of the disordered layers, and the other could be thermal activation at interfaces or direct tunneling of barriers.[7,14]

SIMULATION

On the other hand, the model we use[10] for simulating the photocurrents is based on the following picture: the off-equilibrium charge carriers are represented by a set of particles moving in a random walk across a disordered arrangement of sites, which represents a disordered distribution of localized states. The random arrangement of sites is constructed in a cubic lattice by choosing at random a fraction p of sites, which constitutes a percolation cluster.[15] For this model it has been shown that the fraction of sites that is used to construct the cluster is proportional to the temperature of the system.[16] Because the connectivity of states for the transport is necessary, the fraction p of sites must be always greater than the corresponding percolation threshold p_c for a cubic lattice connecting sites to first nearest neighbors, as establishes percolation theory.[15] Then the average effect of the disordered environment combined with the drift force provided by the applied potential is to highly disperse the off-equilibrium charge packet as it moves across the sample. A complete description of the steps necessary to simulate transient currents in single layers with this model can be found in Refs. 10 and 17 also a good review of numerical simulation of diffusion on fractal-like and percolationlike substrates is given in Ref. 18.

For the case of a multilayer structure, each layer has its own distribution of localized states, drift mobility, etc., according to their own physical properties. Consequently, when a carrier crosses from one layer to other, it undergoes the physical properties of the corresponding layer. However, in this work we assume that the connectivity of localized states remains the same for all the states of the multilayer structure at the same temperature, but the drift velocity and mobility edges have changes going from one layer to other. In this way the total current will be a sum of all the individual photocurrents induced in the complete structure. The latter has been demonstrated in transient photocurrents measured on organic-organic bilayers[5] and in Se-based amorphous multilayer structures.[6] Also, one could consider that the mobility edges are equal or have a mismatch, which results in the formation of a multiple-well (barrier) potential in the sample. These new effects resulting from the barrier formation are introduced in the algorithm by assuming thermal activation with a time distribution of probability of the form

$$P(t)=(1/\tau)\exp(-t/\tau), \tag{2}$$

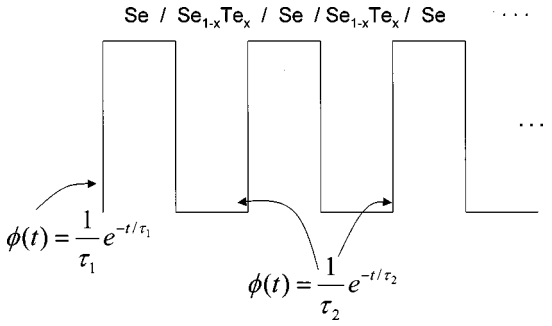

FIG. 1. Representative illustration of the inclusion of the waiting time distribution for activation time in the potential barriers.

with τ the characteristic time at which the thermal activation occurs. As mentioned above, this exponential activation rate is also valid for the thermal release of carriers from single-level traps at the interface. In the algorithm the time delay probability distribution function $P(t)$ is used to evaluate the time a carrier has to wait at an interface before it crosses from one layer to other or from the contact to the first layer. As depicted in Fig. 1, if a carrier moves from a barrier to a valley, then it does not need to be activated, but in the contrary case the $P(t)$ function needs to be evaluated. The characteristic time τ will depend on the kind of barrier the carrier must jump: contact-layer or layer-layer interfaces, which are different. After this step the carrier moves through the disordered media (layer) in order to arrive at the next layer. The process is repeated until the particle reaches the end of the sample where it is absorbed by the contact and ends its contribution to the transient current. Due to the randomness of the disordered media and the waiting times at interfaces, the charge is widely dispersed and the contribution of distant layers is smaller. However, the sum of all contributions from each layer is of considerable magnitude and gives as a result the double-peak-shape photocurrent.

In summary, we have an algorithm for photocurrent simulation that includes two transport mechanisms which could be hopping and thermal activation, the last having two characteristic times τ_1 and τ_2 corresponding to contact-sample and sample-sample interfaces, respectively.

Experimentally, the existence of barriers or deep traps at the interfaces has typical consequences, as is the apparition of single- or double-peak photocurrents. Some details of these effects on the transient photocurrent for a double-layer structure are simulated and compared with experimental data in Ref. 11. Thus we proceed to the comparison of the same effects in multilayer structures.

In order to isolate the influence of each mechanism of transport over the photocurrent shape, we simulated the photocurrent shape for a structure composed of ten cubic layers of width L, with occupation fraction of sites $p\sim 0.52$ and dimension $10L\times L\times L$. First, we simulate (a) the photocurrent shape for a multilayer without barriers at interfaces, neither at contact-sample nor layer-layer interfaces. Then (b) the contact-sample barrier with characteristic activation time τ_c is included, and finally (c) the effects of both contact-sample and layer-layer interfaces with characteristics time τ_c and τ_L, respectively, are included.

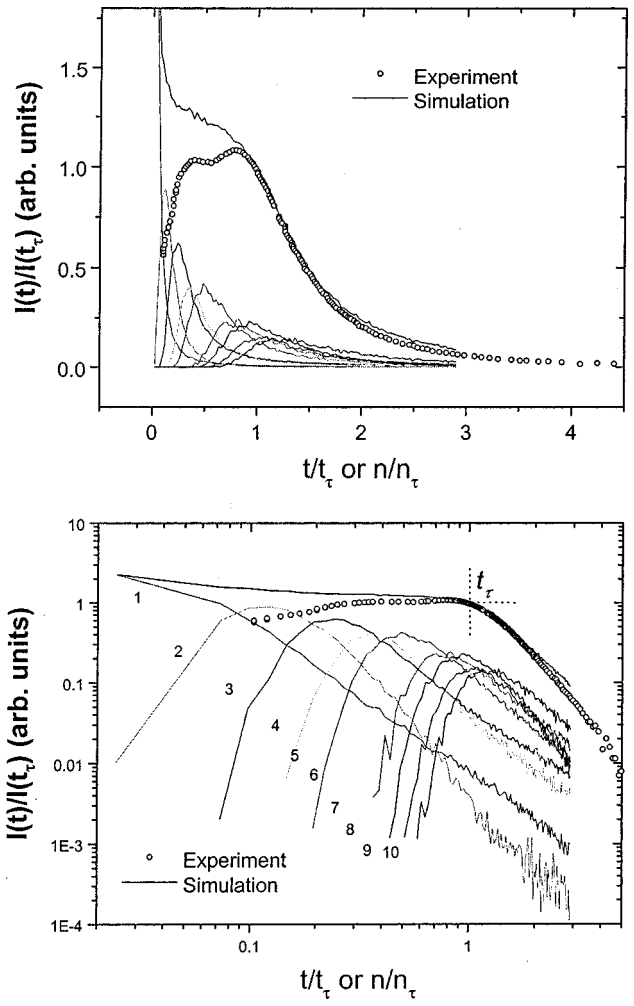

FIG. 2. Experimental and simulated photocurrents without barriers, normalized at the transit time. The same curves on a log–log scale.

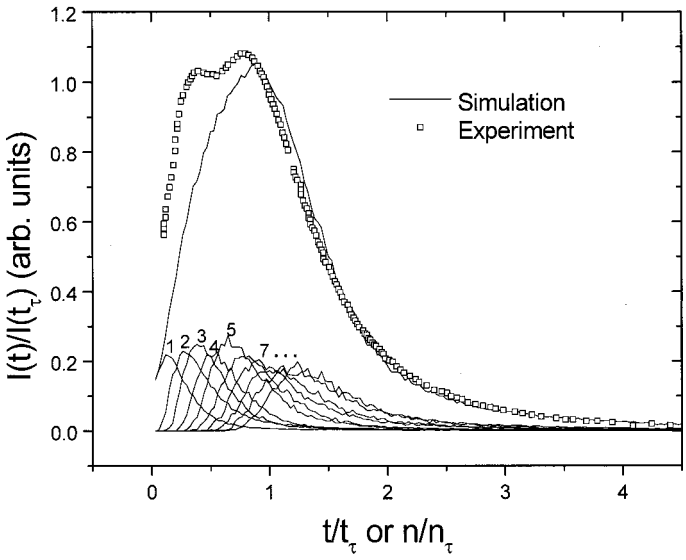

FIG. 3. Fit of experimental and simulated photocurrent including barriers at the contact-sample interface.

RESULTS

Figure 2(a) shows the simulated transient photocurrent for the ten-layer structure without a waiting time function at the contact-layer interface or layer-layer interface. The mobility is the same at any layer, which is equivalent to having one layer 10 times thicker than a single one of the multilayer structure. The total current is the sum of the currents for each single layer, which are enumerated from 1 to 10 in this picture. The dotted line is the experimental results taken from Ref. 13 for $Se/Se_{1-x}Te_x$ multilayers, and both curves are normalized at the transit time t_τ. In Fig. 2(b) the simulated curve is plotted in a logarithmic scale to make clear that both curves coincide at the transit time, which is observed as a change in the slope of the curves as predicted by the CTRW model in Eq. (1). Also, from Fig. 2(a) the current of the first layer behaves like Eq. (1), but none of the $n-1$ currents left have the same behavior; instead, all they have the form of peak due to the natural time delay of carriers produced by the dispersion at crossing the previous layer.

For the case when the time delay of charge at the contact-sample interface where the off-equilibrium carriers are injected is included, then the photocurrent will start at zero at $t=0$, as is shown in Fig. 3. The photocurrent now starts from zero intensity, but still the double-peak-shape is missing, as in the experimental case of the dotted curve. The contributions from each layer are marked from 1 to 10, and all have the same one-peak shape as expected for a single layer with time-delayed injected charge.[10] Also, the amplitudes of each curve are approximately in the same order because the charge carriers flow without delay at the interfaces of the multilayers structure.

Hence the next natural step is to include a mismatch mobility edge at each interface by considering a time delay function at a position where the mobility edge changes from a lower to a higher value, keeping the time delay distribution function at the contact-sample interface. These results are presented in Fig. 4, from where one can see that the photocurrent shape is better approximated to the experimental data from Ref. 13. The contributions of each layer to the total current clearly show that the first peak of the total current is due mainly to the current induced in the first three layers and controlled by the time delay introduced by the contact-sample interface. The sum of the small contributions from the rest of the layers gives form to the second peak of the total photocurrent. Figure 4(b) shows a composition of the total current as a sum of two peaks originating from the sum of the first three currents and the currents left. There is no doubt of the roll played for the time delay functions at the interfaces and that the peak shape of the photocurrents is inherently linked to it. One important point is that our proposed mechanism by means of which delayed charge carriers cross interfaces holds for this particular case of multilayers structure. Furthermore, our simulation results are compatible with a thermal activation mechanism of carriers over a well-defined energy barrier, which height defines the characteristic waiting time at interfaces.

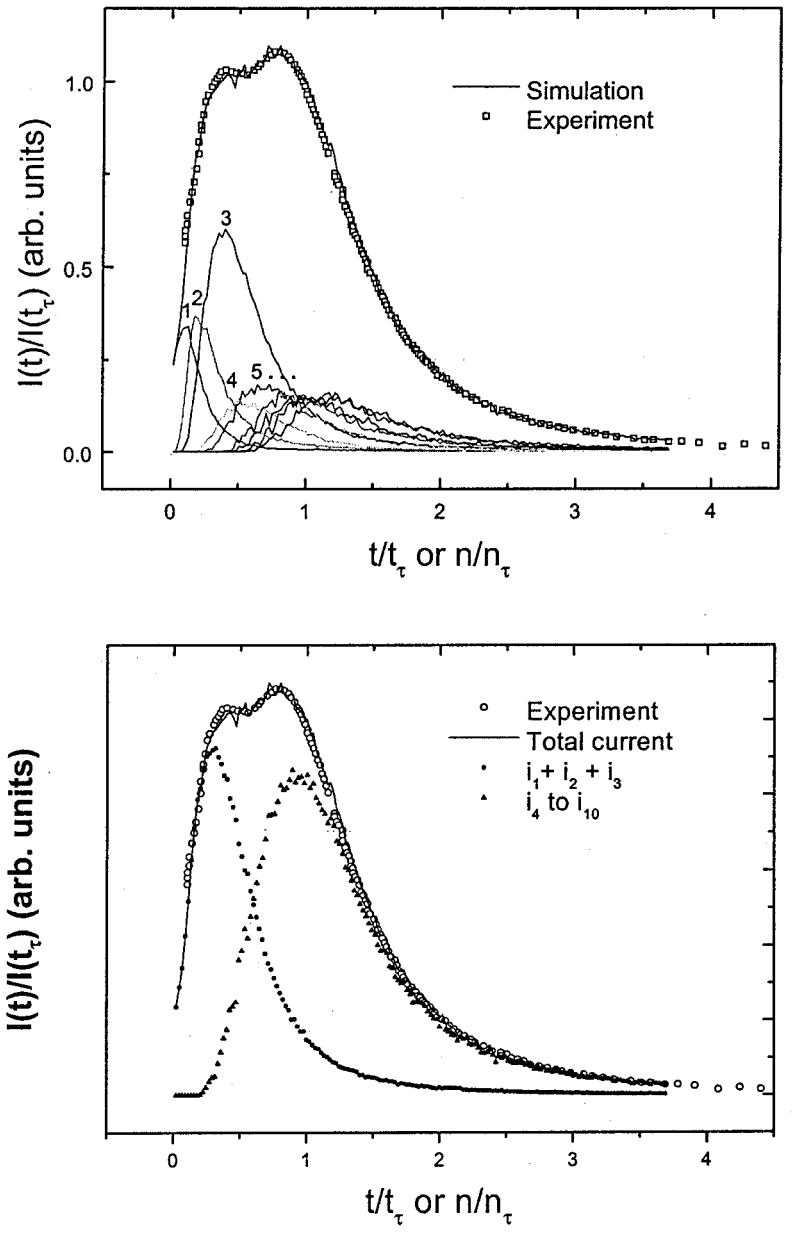

FIG. 4. Fit of experimental and simulated photocurrent including waiting time distribution functions at the barrier interfaces: contributions from each layer and contributions from layers 1–3 and 4–10, respectively.

Thus the first peak of the photocurrent depends on the characteristics of the first three layers, and the second peak depends on the subsequent $n-3$ layers of the multilayer structure. The entity responsible for this behavior is the contact-sample interface by means of the time-dependent probability assigned to it. The time-delayed charge carriers injected from this interface have effects on the current of the next layers, as is clearly evident on the first two components of the total current in Fig. 4(a). These peaks are sharp and ''unfinished;'' i.e., the carriers move to the next layer before all they are injected, which means that the time needed for a carrier to cross an interface without barrier is less than the time needed to be activated at the interface. In the third component of the same figure, the induced current is larger due to the barrier existing at the layer-2–layer-3 interface. An equilibration of times occurs here for the carriers that are arriving at this interface and those that are waiting to be activated. It means that the times at which carriers cross from layer 1 or layer 2 to layer 3 are contained in a range of time which coincides with the time of activation at this place, and consequently the number of carriers moving inside the layer 3 is larger than for any other layer. The same can occur for the first or second layer depending on the transit time and the layer thickness. This effect on the photocurrent is supported by the experimental results of Ref. 13, that the first-peak behavior of $Se/Se_{1-x}Te_x$ multilayers with electric field strength E and temperature T is similar to that of a single $Se_{1-x}Te_x$ layer.

Looking for more experimental evidence to prove our ideas, we simulated the two-peak to one-peak transformation for the photocurrent when the layer thickness is decreased, maintaining the same characteristic time. Using different sizes of layer lengths and keeping constant the electric field and temperature, a series of currents were simulated. Figure

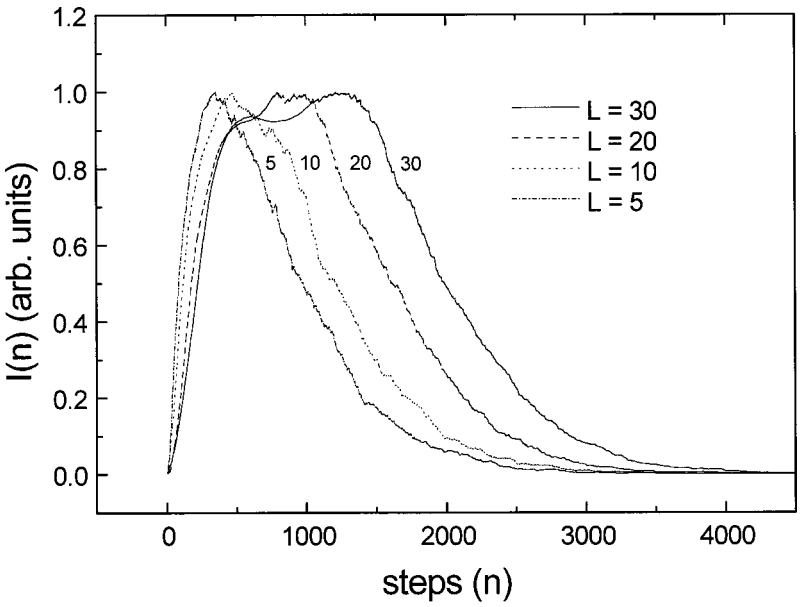

FIG. 5. Behavior of simulated photocurrent with variation of the layer thickness from $L=5$ to 30.

5 shows how as L is decreased the double-peak appearance of photocurrent is turned into an one-peak shape curve. The same results were found by other authors[13] by measuring TOF photocurrents for $Se/Se_{1-x}Te_x$ multilayers. They found that going from $[250/250]\times 50$ Å thickness multilayers to $[100/50]\times 75$ Å thickness multilayers a two-peak to one-peak transition is carried out.

CONCLUSIONS

Finally, we conclude that time delay of carriers at interfaces contribute to slowing down the carrier movement, but it is the disordered distribution of sites which contributes to the decaying current slope. We identified thermal activation as the mechanism that allows charge carriers to cross over the interface barriers.

The peak shape of photocurrent is due to the existence of a time delay of carriers at interfaces, combined with a hopping mechanism across the disordered sample. In our system the first peak results mainly from the competence between the activation of carriers at the contact-sample and layer-2–layer-3 interfaces, which accumulates more carriers inside a layer in a period of time. The second peak is due to the contribution of the currents induced in the rest of layers as seen in Fig. 4(b). The good correspondence of the simulated results on a 10-layer structure to a 100-layer system is due uniquely to the character of the self-similarity of the transient transport, for which the transit time is imposed by the system thickness.

Also, when the layer thickness is lowered in the multilayer structure, the two-peak shape of photocurrent can transform into a one-peak shape. This occurs by the coupling of waiting time distributions at interfaces by the reduction of the time needed by a carrier to cross a single layer, keeping the transport mechanism unaltered.

ACKNOWLEDGMENT

This work was partially supported by Consejo Nacional de Ciencia y Tecnología (CONACyT-México).

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