Multiple trapping in strong electric fields

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The time-dependent and steady-state drift mobilities of photoexcited carriers are calculated within a framework of a multiple-trapping model incorporating nonlinear effects due to a strong electric field. Tunneling processes from localized states to the transport edge and vice versa are taken into account. We discuss the application of the proposed theory to recent experiments and compare it with hopping theory.

INTRODUCTION

Recent experiments on the electric-field-dependent electron transport in amorphous Si:H (Refs. 1 and 2) have made available both time-dependent and steady-state data, which have shown that the nonlinear regime is interesting and must be compared with existing transport models.

A quantitative theory developed by Scher and Lax,[3] and Scher and Montroll,[3] the so-called theory of continuous-time random walk (CTRW), showed in general how to replace the random system by an ordered system (with respect to the transfer rates) with a selected "waiting-time distribution." This theory has been used extensively to describe the time-dependent transport properties (dispersion) of disordered materials. This theory can be reformulated as an effective-medium approximation, and Schmidlin[4] argued that the multiple-trapping (MT) process leads to a dispersion equivalent to that of CTRW. In this approach an electron may be trapped to a localized site, and then excited above the mobility edge due to thermal excitations. The electron is mobile only above the mobility edge and the averaged mobility is proportional to the number of excited electrons. Tiedje[5] and Hvam and Brodsky[6] showed that MT can account for the transport properties of amorphous Si:H, invoking an exponential approximation for the band-tail density of states.

Although the MT description is adequate in the temperature range 90–400 K, at lower temperatures the hopping among the localized states has been taken into account. Nevertheless, due to the work by Silver, Schönherr and Bässler[7] followed by other works, we know that hopping might provide dispersion analogous to that of MT. The existing theory, which includes hopping as presented by Grünewald *et al.*,[8] requires computer assistance and so does its extension to nonlinear regimes.[9] It became especially clear that hopping can be understood as a MT process when Monroe[10] emphasized that the *transport* energy plays the role of the "mobility edge." It is reasonable, therefore, to apply the MT theory to more advanced problems, such as high-field mobility (which is discussed here), magnetic properties, etc. Semiquantitative arguments are not universal, and MT theory is not a universal approximation; it requires careful checks for each application. However, we believe that the temperature interval 90–400 K can be quantitatively described (see our theoretical fit below). The MT approach is simple and provides effective description for experimental needs. It is useful to compare the high-field MT predictions with the "effective temperature" conjecture of the hopping model proposed by Shklovskii *et al.*[11]

The classical MT mobility does not depend upon electric field. One possibility of taking the field into account is to consider the *tunneling* from localizing traps to the transport edge in the field direction (see Fig. 1). Of course, at high enough temperatures this process has to be activation assisted, when an optimal excitation and tunneling are combined, but we deal here only with the simplest case to demonstrate the model. The detailed balance requires an account for the opposite process of tunnel trapping.

The paper is organized as follows. In the following section we solve the rate equations for MT in the presence of tunneling to the transport edge and vice versa. For the time-of-flight experiments the time-dependent current is calculated for the times before and after the transit time,[5] which is the average drift time. The steady-state current is also obtained. In the subsequent section we compare our model with experimental results.[1,2]

THE MT RATE EQUATIONS WITH TUNNELING

We follow the Tiedje treatment of multiple trapping[5] and investigate the linear system

$$\begin{aligned}
&\frac{\partial n}{\partial t}+\frac{\partial}{\partial x}\left[\mu_0 nE+\sum_j (\Delta x)_j R_j(E) n_j\right]=\sum_j \{[r_j+R_j(E)]n_j-[c_j+C_j(E)]n\}+\delta(x)\delta(t)\;,\\
&\frac{\partial n_j}{\partial t}-(\Delta x)_j C_j(E)\frac{\partial n}{\partial x}=-\{[r_j+R_j(E)]n_j-[c_j+C_j(E)]n\}\;.
\end{aligned}\qquad(1)$$

Here $n(x,t)$, and $n_j(x,t)$ describe free and trapped electrons. Subscript j numerates all traps. The left-hand term with the sum represents the current caused by tunneling (E argument). The right-hand sum describes trapping and thermal excitation. We select the energy axis directed downwards (see Fig. 1). Here r_j is the MT "release" rate constant, which equals

$$r_j=\nu_0 e^{-\varepsilon/T} , \tag{2}$$

and c_j describes the "capture" rate constant

$$c_j=\nu_0 g(\varepsilon) , \tag{3}$$

where the continuum limit is used. The constants (2) and (3) satisfy the detailed balance condition, and the density of states $g(\varepsilon)$ is

$$g(\varepsilon)=(1/\varepsilon_0)e^{-\varepsilon/\varepsilon_0} . \tag{4}$$

The tunneling constant of release is

$$R_j(E)=\nu_0 e^{-2(\Delta x_j)/a}=\nu_0 e^{-2\varepsilon/eaE} , \tag{5}$$

and that of capture equals

$$C_j(E)=\nu_0 g(\varepsilon)e^{-2\varepsilon/eaE} ; \tag{6}$$

these rate constants are also related by the detailed balance.

Note, that the tunneling current in (1) is distributed between two equations. To clarify this distribution one may consider a lattice model. For example, the income tunneling term for a trap population is $C_j(E)n[x+(\Delta x)_j]$, which gives the current upon expanding.

The system (1)–(6) may be solved by a Fourier *time* transform, if we are interested in drift properties only. It means that we actually omit all the terms which are proportional to the squared wave vector, or E^2. The resulting equation for the image $\tilde{n}(\omega,x)$ is

$$i\omega\tilde{n}+[\mu_0\tilde{n}E+\tilde{v}(\omega,E)]\frac{\partial\tilde{n}}{\partial x}=-i\omega K(\omega,E)\tilde{n}+\delta(x) , \tag{7}$$

with the tunneling "velocity"

$$\tilde{v}(\omega,E)=\sum_j \frac{R_j(E)c_j-C_j(E)r_j}{i\omega+r_j+R_j(E)}(\Delta x)_j , \tag{8}$$

and the combined rate constant,

$$K(\omega,E)=\sum_j \frac{c_j+C_j(E)}{i\omega+r_j+R_j(E)} . \tag{9}$$

All the sums over localized states may be replaced by integrals, using the simple rule $\sum_j=\int d\varepsilon$. The substitution $\varepsilon=-T\ln u$ is used below.

We first evaluate integrals for Eqs. (8) and (9). The tunneling velocity from Eq. (8) may be written as follows:

$$\frac{\tilde{v}(\omega,E)}{\mu_0 E}=\frac{e\nu_0 a^2}{3\mu_0 T}f(\alpha,\beta) ,$$
$$f(\alpha,\beta)=\tfrac{3}{4}\alpha\beta^2\int_0^1 du\frac{u^{\alpha+\beta-1}(1-u)}{\dfrac{i\omega}{\nu_0}+u+u^\beta}\ln\left[\frac{1}{u}\right] , \tag{8a}$$

with the usual MT parameter $\alpha=T/\varepsilon_0$, and a new field-dependent one $\beta=2T/eaE$; compare their ratio with the definition of Shklovskii *et al.* of the effective temperature in the strong electric field.[11] Integral (8a) gains its value in the region $u\sim 1$. Therefore one can definitely omit the frequency-dependent term in the denominator, as soon as $\omega/\nu_0\ll 1$ at the times of relevance; ν_0 has an atomic order. It means that the tunneling velocity, or nonlinear input in mobility, is frequency independent.

Integral (8a) can be calculated exactly,

$$f(\alpha,\beta)=\begin{cases}\dfrac{3\alpha\beta^2}{8(\beta-1)^2}\left[F\left[\dfrac{\beta+\alpha-1}{\beta-1}\right]-F\left[\dfrac{\beta+\alpha}{\beta-1}\right]\right], & \beta>1 ,\\[2ex] \dfrac{3\alpha\beta^2}{8(\beta-1)^2}\left[F\left[\dfrac{\alpha}{1-\beta}\right]-F\left[\dfrac{\alpha+1}{1-\beta}\right]\right], & \beta<1 ,\\[2ex] \dfrac{3(2\alpha+1)}{8\alpha(\alpha+1)^2}, & \beta=1 .\end{cases}$$
$$F(z)=\psi^{(1)}\left[\frac{z+1}{2}\right]-\psi^{(1)}\left[\frac{z}{2}\right] . \tag{8b}$$

Here $\psi^{(1)}$ is the trigamma function. In low fields, $\beta\gg 1$, the asymptotic form is valid, $f(\alpha,\beta)=\frac{9}{2}\zeta(3)\alpha\beta^{-1}$, so that the field-dependent mobility behaves like $|E|$, $[\mu(E)-\mu_0]\propto|E|$. At large fields, in the limit $\alpha,\beta\ll 1$, the asymptotic form is $f(\alpha,\beta)=3\alpha\beta^2/(\alpha+\beta)^2$. Therefore, strong electric field first increases and then decreases the effective mobility almost back to the zero-field value, μ_0. For a given temperature (fixed α) the maximum is at $\beta\gtrsim 1$, and its value is close to the $f(\alpha,1)$ listed above. It means that there exists a maximum nonlinearity of the mobility at the field $E\sim 2T/ae$. The nonlinear part of the mobility can be large.

Now we evaluate the combined rate constant

$$K(\omega,E)=\alpha\int_0^1 du\frac{u^{\alpha-1}+u^{\beta-1}}{\dfrac{i\omega}{\nu_0}+u+u^\beta} . \tag{9a}$$

The value of this integral also depends upon electric field in a curious manner. At fields when $\beta > 1$ (note the "$>$," not "$\gg$," the precision is exponential), one obtains the regular MT result,

$$K(\omega)=\frac{\alpha\pi\left[\dfrac{i\omega}{\nu_0}\right]^{\alpha-1}}{\sin\alpha\pi}\ . \tag{9b}$$

Because the field dependence is absent, all the expressions in this case are factorized in field- and time-dependent multipliers (as was measured in Ref. 2). It means that energy and space evolutions are independent. Electrons at a given point in space are rapidly distributed along the energy axis in accordance with the time available. The independent process is the tunneling current, which occurs slowly; but the tunneling length is large and makes it possible to dominate the mobility.

In the case $\alpha < \beta < 1$, the field enters the dispersion parameter:

$$K(\omega,E)=\frac{\alpha\pi\left[\dfrac{i\omega}{\nu_0}\right]^{\alpha/\beta-1}}{\sin\left[\dfrac{\alpha}{\beta}\pi\right]}\ . \tag{9c}$$

Therefore, this field range supports the conjecture of Shklovskii *et al.* of the hopping approach; the ratio α/β would be a dispersion parameter if $eaE/2$ is believed to substitute for the temperature.[11] Note, that the "transition" from the regular temperature to this "effective" temperature can be summarized as

$$T_{\text{eff}}=\max(T,eaE/2)\ . \tag{10}$$

The remaining case of even stronger fields, $\beta < \alpha$ is different:

$$K(\omega,E)=\frac{\alpha}{\beta}\ln\left[\frac{i\omega}{\nu_0}\right]\ . \tag{9d}$$

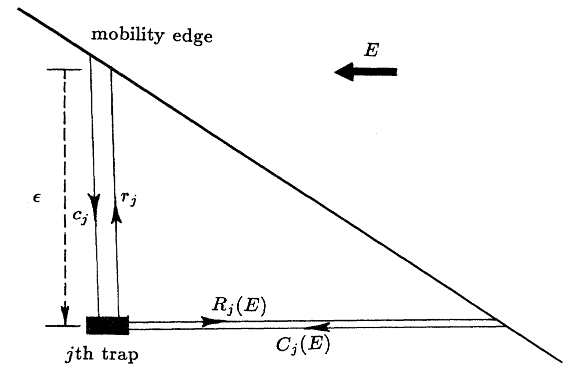

FIG. 1. The kinetic processes taken into account in the proposed model. Two vertical lines indexed by c_j and r_j represent regular multiple traping; and two horizontal lines with $C_j(E)$ and $R_j(E)$ show the tunneling for the jth trap.

Now we may integrate Eq. (7) and find the inverse transform.

Note that some of the features are sensitive to the model assumptions. Namely, specific behavior in the vicinity of the points, $\beta=1$, $\beta=\alpha$, i.e., the different laws coexisting with the exponential precision, is an artifact which may be smeared by more realistic approaches. One may adopt, for example, that the localization radius is energy dependent, $a(\varepsilon)$.[11] We shall not discuss this in detail. The nonanalyticity $[\mu(E)-\mu_0]\propto|E|$ which was found in the low-field limit is also a by-product of *one-side* tunneling (see Fig. 1). However, in the presence of hopping, both-sides jumps provide the proper mobility at low fields, $[\mu(E)-\mu_0]\propto E^2$.

The quantity which is measured in Refs. 1 and 2 is the current, which is defined as

$$\begin{aligned}I(E,t)&=\frac{\mu_0E+\bar{v}(E)}{L}\int_0^L n(x,t)dx\\&=\frac{\mu_0E+\bar{v}(E)}{2\pi L}\int_{-\infty}^{\infty}d\omega\frac{e^{i\omega t}}{i\omega[1+K(\omega,E)]}\left[1-\exp\left[-\frac{i\omega L[1+K(\omega,E)]}{\mu_0E+\bar{v}(E)}\right]\right]\ .\end{aligned} \tag{11}$$

TABLE I. Electric current; asymptotic expressions.

	$t \gg \tau_T$	$t \ll \tau_T$
$\beta>1$	$\dfrac{\Gamma(1+\alpha)\alpha\nu_0^2L}{2(\mu_0E+\bar{v})(\nu_0t)^{1+\alpha}}$	$\dfrac{\Gamma(1-\alpha)\sin^2(\alpha\pi)(\mu_0E+\bar{v})}{\alpha\pi^2L(\nu_0t)^{1-\alpha}}$
$1>\beta>\alpha$	$\dfrac{\Gamma(1+\alpha/\beta)\alpha\nu_0^2L}{2(\mu_0E+\bar{v})(\nu_0t)^{1+\alpha/\beta}}$	$\dfrac{\Gamma(1-\alpha/\beta)\beta^2\sin^2(\alpha/\beta\pi)(\mu_0E+\bar{v})}{\alpha\pi^2L(\nu_0t)^{1-\alpha/\beta}}$
$\beta<\alpha$	$\dfrac{\alpha L}{2\beta t^2(\mu_0E+\bar{v})}$	$\dfrac{\beta(\mu_0E+\bar{v})}{\alpha L\ln(1/\nu_0t)}$

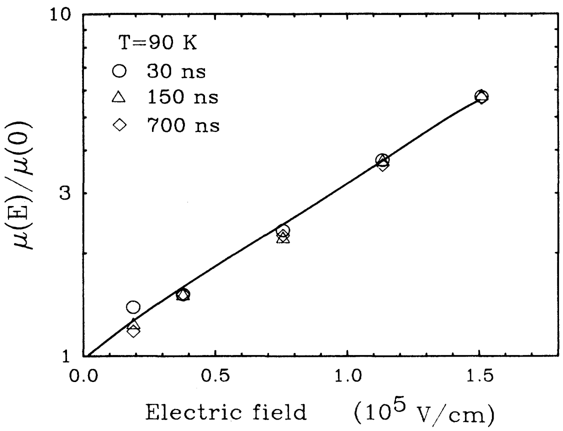

FIG. 2. Figure 10(a) of Ref. 2 is fitted by Eqs. (8a), (8b), and (13). Parameters taken from Ref. 2: $T=90$ K, $\varepsilon_0=257$ K, $\mu_0=1.0\ \mathrm{cm^2/V\,s}$. Parameter used, $\nu_0 a^2=0.12\ \mathrm{cm^2/s}$.

In the steady-state excitation experiments[1] we assumed that the recombination time τ_R does not depend on field and is small with respect to transit time, so that the concentration n is uniform and constant. We use this oversimplified assumption to focus on the mobility behavior only. Then the current equals

$$I(E)=[\mu_0 E+\bar{v}(E)]n\ . \tag{12}$$

For the current in the time-of-flight experiments there are two time limits with respect to transit time and three field intervals, $\beta>1$, $1>\beta>\alpha$, $\alpha>\beta$. The results for the current in these six cases are collected in Table I. There τ_T means the transit time which is estimated as the time where two asymptotical answers match.

COMPARISON WITH EXPERIMENTS (REFS. 1 AND 2)

Let us begin with Ref. 2, where we found a quantitative accordance using one adjustable parameter. Antoniadis and Schiff have interpreted their data as a proposal of the field and time factorized dependencies [see Fig. 6 and Eq. (9) of Ref. 2]. This is consistent with Table I in the regime before the transit time. We found it also possible to fit their Fig. 10(a) using the formula

$$\frac{\mu(E)}{\mu_0}=1+\frac{\bar{v}(E)}{\mu_0 E} \tag{13}$$

together with Eqs. (8a) and (8b), and parameters taken from Ref. 2, $T=90$ K, $\varepsilon_0=257$ K ($\alpha=0.35$), $\mu_0=1.0\ \mathrm{cm^2/V\,s}$. The result is shown in Fig. 2. The fit could be

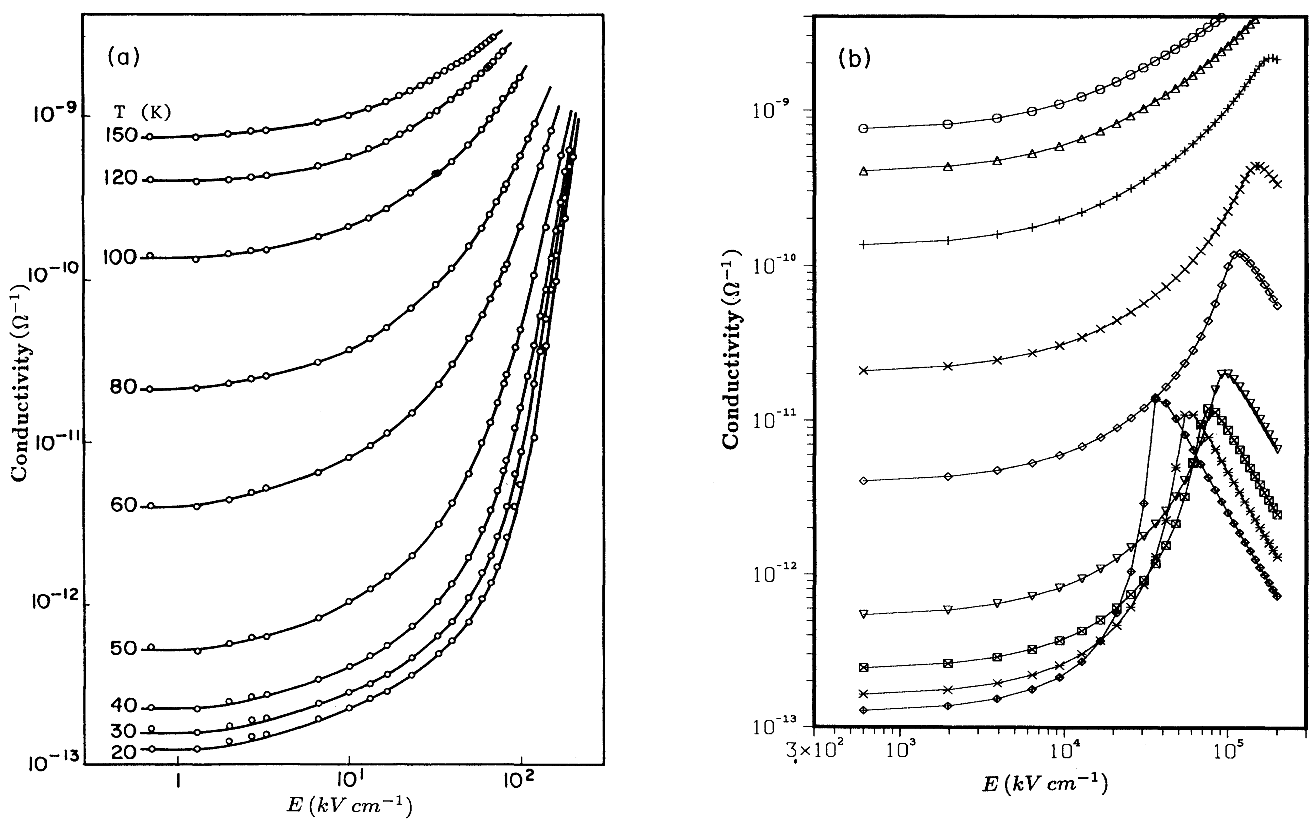

FIG. 3. Figure 4 by Stachowitz, Fuhs, and Jahn (Ref. 1) (a) is fitted by Eqs. (8a), (8b), and (12) (b). Parameters taken from Ref. 1: zero-field conductivities to normalize the curves. Previously selected values: $\varepsilon_0=257$ K, $\mu_0=1.0\ \mathrm{cm^2/V\,s}$. From the fit: $\nu_0=4.0\times10^{13}\ \mathrm{s^{-1}}$, $a=10$ Å.

done at any point in the (ν_0, a) plane, along the line $\nu_0 a^2 = 0.12\ \text{cm}^2/\text{s}$, if $a < 10$ Å. We preferred to select the smallest ν_0, so it follows that $\nu_0 = 1.2 \times 10^{13}$ s^{-1}, $a = 10$ Å, which seems to be reasonable.

According to Eq. (12) we may try the same formula for the data of Stachowitz, Fuhs, and Jahn.[1] Because the concentration n is not known, we fix the low-field conductances for different temperatures using Figs. 1 and 4 of Ref. 1. To the best of Eq. (12) possibilities we were able to fit high-temperature curves (100, 120, and 150 K), selecting $\nu_0 = 4.0 \times 10^{13}\ \text{s}^{-1}$, $a = 10$ Å. The low-temperature curves cannot be described, because the nonlinearity in our approach is restricted [see (8b)]. It means that the transport energy and concentration do *change* with field; this effect lies outside the exploited model. The fit could be improved if we take into account the field dependence of the recombination time.[11]

CONCLUSION

We proposed a simple model of multiple trapping with the tunneling to the transport edge and vice versa in the strong electric field. At temperatures $T > 90$ K this model may be used for experimental fitting. We have also shown how to connect the temperature and the high-field effective temperature, proposed in Ref. 11.

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