Monte Carlo Calculation of the Transient Photocurrent in Low-Carrier-Mobility Materials*

M. Silver, K. S. Dy, and I. L. Huang
University of North Carolina, Chapel Hill, North Carolina 27514
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A Monte Carlo method is used to calculate the shape of the transient photocurrent under flash excitation. It is shown that some of the anomalous features of the experimental data can be explained by including surface as well as bulk trapping in the model. In particular, this simple picture predicts the observed long tail of the pulse as well as the apparent field dependence of the mobility.

In recent years transient photocurrents have been examined very successfully in low-mobility material. Our present day understanding of the electronic properties of organic[1] and molecular[2] crystals and amorphous semiconductors[3] results largely from interpretation of such experiments. In addition to obtaining information on drift mobilities, data on trapping cross sections were also obtained.

However, one problem persisted: The theoretical pulse shape was not always observed even in the same material. Typical photocurrents are shown in Fig. 1. We show in Fig. 1(a) a curve obtained from anthracene, which closely resembles the theoretical pulse shape. Figure 1(b) shows another pulse obtained from anthracene with a different electrode. Finally, Fig. 1(c) shows a typical pulse obtained from As_2Se_3. Noticeable

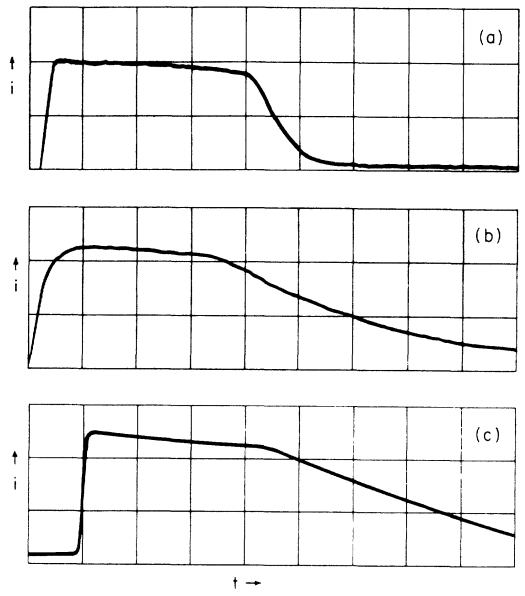

FIG. 1. Typical transient current pulses excited by weak-intensity, strongly absorbed light incident upon (a), (b) anthracene and (c) As_2Se_3. The curve in (c) is taken from Fig. 5 of Ref. (3).

in Figs. 1(b) and 1(c) are very long tails after a break in the curve which ostensibly represents the transit time.

The existence of this long tail prompted Scharfe[4] to suggest that it was due to a dispersion in the arrival time of the carriers due to a distribution of mobilities. This interpretation seemed physically unsound because it implies that different carriers sample different densities of states or different traps. This would be equivalent to having parallel strips of different materials.

We propose a different and physically more sound model: The generated carriers, in addition to being trapped in the bulk, may also be trapped at the illuminated surface. The surface trapping is equivalent to a delay in the carrier generation.

In order to illustrate that such a simple process can give the desired pulse shape, we further simplify the model by assuming that there is only one trapping level in the bulk and only one level at the surface. It is not significantly more difficult to include a distribution of levels and even surface recombination. (Calculations including these cases are presently being undertaken.)

In order to calculate the current pulse we have chosen to use a simplified Monte Carlo approach rather than solve the continuity of current equation. We took this approach because we wanted to make as few mathematical assumptions and approximations as possible.

In the Monte Carlo approach we only have to choose a trapping probability and an escape probability. We do not have to be concerned with equilibrium between the trapping states and the conduction band at any time from $t=0$ to $t=\infty$. Any approach to equilibrium will automatically come about through the statistics.

This calculation is extraordinarily simple. One first chooses a surface-trapping probability; this is just $N_D/(N_D+N_I)$, where N_D is the number of carriers trapped at the surface and N_D+N_I is

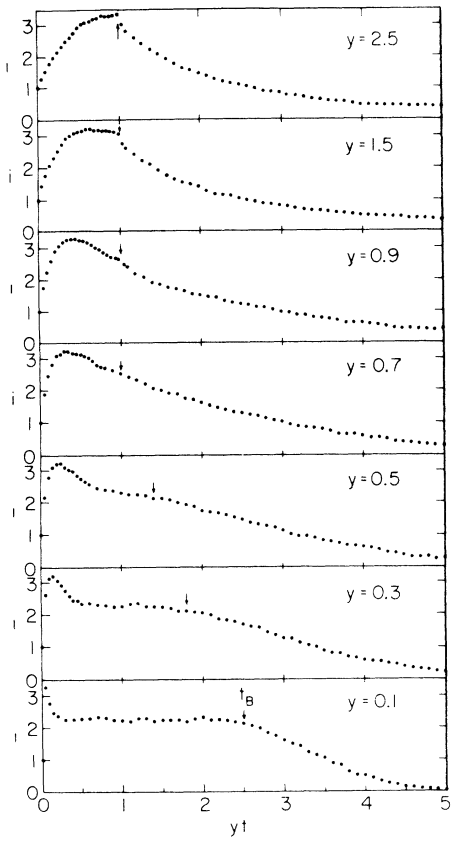

FIG. 2. Plot of the current versus time multiplied by the voltage parameter y obtained from a Monte Carlo calculation for a system with $N_I = 1000$, $N_D = 7000$, $\tau_\alpha = \tau_\gamma = 4 \times 10^{-4}$, $\tau_\beta = 10^{-3}$, and $t_0 = 10^{-3}$. The yt axis is in units of t_0.

the total number of carriers.

The N_I carriers are instantaneously injected into the bulk, and using a set of random numbers we can follow the history of each one. The sequence of events are as follows:

first trapping time, $t_T(1) = -\tau_\alpha \ln r_T(1)$;

first emptying time, $t_e(1) = -\tau_\beta \ln r_e(1)$;

...

nth trapping time, $t_T(n) = -\tau_\alpha \ln r_T(n)$;

nth emptying time, $t_e(n) = -\tau_\beta \ln r_e(n)$,

where τ_α and τ_β are the characteristic trapping and emptying time, respectively, for bulk traps. This sequence is continued until

$$\sum_{n=1}^{m} t_T(n) = t_0/y,$$

where t_0/y is the transit time (y is a parameter which plays the role of the applied voltage, and t_0 is a fixed constant). The trapping time for the final event $t_T(m)$ usually has to be cut short to make the sum exactly equal to t_0/y. The values of $r_T(n)$ and $r_e(n)$ are random numbers generated by a computer. By following each particle we can tell at any time how many are free, and the sum of these is proportional to the current.

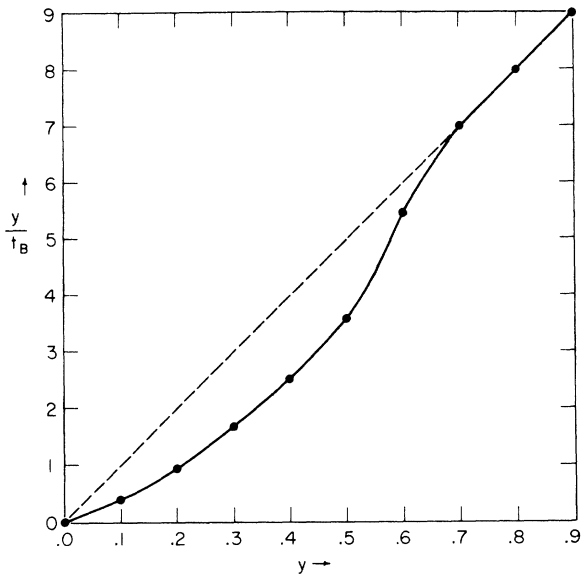

FIG. 3. Plot of y/t_B vs y, where t_B is determined from the break in the current pulses as shown in Fig. 2.

The procedure for the N_D particles is the same as for the N_I particles except that the first event is a surface-trap-emptying one with a distribution of emptying times given by $t_s(1) = -\tau_\gamma \ln r_s(1)$, τ_γ being the characteristic release time for surface traps. Figure 2 shows a set of the results for the number of free carriers at a given time t plotted against yt for $N_I = 1000$, $N_D = 7000$, and $y = 0.1$ through 2.5. In all cases, we have chosen the values $\tau_\alpha = \tau_\gamma = 4 \times 10^{-4}$ sec, $\tau_\beta = 10^{-3}$ sec, and $t_0 = 10^{-3}$ sec. We see that only at high voltages does one get a break in the curve at $yt = t_0$. Further, it had previously been assumed that by measuring the decay of the current before the break, one could obtain information on the trapping time. As can be seen, neither t_0 nor τ_α are derivable by such simple procedures as observing the decay and the break point.

Finally, one can plot the reciprocal of the apparent transit time (the break point t_B) versus the applied voltage. These results are shown in Fig. 3. There is an apparent field-dependent mobility which is nothing more than a transition from the trap-controlled to the trap-free case with increasing voltage. This curve is very similar in shape to that obtained by Tabak[3] who pos-

tulated a field-dependent mobility to explain the results.

We conclude from these calculations that the so-called complicated drift experimental results obtained on amorphous materials such as As_2Se_3 may in fact be very simple and involve well-known processes. Further, to derive the mobility and the trapping times one must study the current pulses in detail as a function of field and temperature.

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Evidence of Strong Optical Super-Radiant Damping in Ruby*

A. Compaan and I. D. Abella
Department of Physics, University of Chicago, Chicago, Illinois 60637
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We report measurements of direct photon-echo intensity versus Cr concentration which indicate the presence of sizable radiation-damping effects in dark ruby. The departure from n^2 dependence for concentrations above 0.03% Cr demonstrates that modifications of self-induced transparency analyses are necessary in dense resonant media.

We report results of photon-echo[1] experiments which indicate, for the first time, the occurrence of sizable radiation damping of the super-radiant state[2] formed by the initial excitation pulse. This follows from our observation of the failure of the predicted n^2 concentration dependence of the echo intensity at concentrations above ~0.03%. We find the radiation-damping lifetime of the initial state in high-chromium-concentration ruby to be of the same order as, or less than, the incident pulse width $\Delta\tau$ of 15 nsec. We shall show that these results imply that modifications are necessary in the analyses[3,4] of resonant-pulse propagation in dense media.

Photon echoes result from the enhanced radiation rate of the reconstructed super-radiant state, at $t=2\tau$, in response to excitation pulses at $t=0$ and $t=\tau$. Although much smaller than the spontaneous emission lifetime of a single atom, the super-radiant damping lifetime defined below is usually large compared to the Bloch decay time for most systems studied so far,[5] and the damping is safely neglected. Radiation damping in spin systems has been discussed by several authors,[6,7] with times as short as 10^{-7} sec expected in the microwave region for spin densities of ~10^{17} cm^{-3} and high-Q cavities. Although radiative damping is expected to increase as ω^3, weak matrix elements in the case of ruby and the general effect[1] of volumes larger than λ^3 contribute to negligible damping at optical frequencies for dilute systems.

The experiments we report here were performed on ruby samples of various Cr concentrations at $T=2.2$°K, and we shall now indicate briefly the connection between the photon-echo measurement at $t=2\tau$ and the super-radiant decay at $t=0$. One may define[7-9] a super-radiant lifetime T_s in terms of which we write the super-radiant emission which immediately follows the first incident pulse as

$$I_s=(N\hbar\omega/T_s)|\langle\exp(-i\Delta\omega t)\rangle_{\mathrm{av}}|^2\sin^2\theta,$$

where

$$T_s^{-1}=\tfrac{1}{4}N\int d\Omega\, T_1^{-1}|\langle\exp[-i(\vec{k}-\vec{k}_1)\cdot\vec{r}]\rangle_{\mathrm{av}}|^2.$$

For large Fresnel number[10] the integral over all directions $\hat{k}$ gives $T_s^{-1}=N\lambda^2/4T_1A=n'l\lambda^2/4T_1$. N and n' are, respectively, the total number and density of echo-producing ions; the pulse "area" θ is given by $\theta=(p/\hbar)\int_0^{\Delta\tau}\mathcal{E}dt$, where p is the dipole matrix element of the transition and $\mathcal{E}$ is the envelope of the electron field with wave vector in the medium of $|\vec{k}_1|=2\pi/\lambda$. T_1 is the spontaneous-emission lifetime of a single atom, $\Delta\omega$ is the difference between the transition frequency of a given atom and the frequency of the incident laser field, and $\vec{r}$ denotes the position of that atom; A is the cross-sectional area of the incident beam in the crystal and l is the crystal thickness. The indicated averages are taken over all ions participating in the echo. The factor $|\langle\exp(-i\Delta\omega t)\rangle_{\mathrm{av}}|^2$