in this material. Therefore, the strong field dependence of the apparent injection efficiency is not a unique characteristic of the space-charge-limited current, but is characteristic of measurements made while a transit is still proceeding. A typical xerographic apparatus response time is ~0.1 sec and, referring to Fig. 4, it is seen that the transit time of this PVK sample already begins to exceed this value at fields $\sim 3\times10^4$ V/cm. The transition referred to above is, therefore, expected to occur in the range of field values necessary to account for the observed xerographic gain-vs-field curves. This bulk limitation does not arise from any permanent loss of carriers, but from the fact that, because of their low velocity, the carriers have not sufficient time to transit the PVK before the initial dv/dt is measured. In a xerographic sense, the usual meaning of a bulk limitation is a range limitation, i.e., $\mu\tau E <$ sample thickness, where τ is the bulk deep-trapping lifetime. It is now clea that another bulk limitation can arise if the time required to transport the charge is inconveniently long. By an arbitrary cutoff in the time of observation, this will be an operational range limitation, and the xerographic gain at a given field will be inversely proportional to the PVK thickness. Colloquially stated, it matters how fast the carriers move as well as how far.

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Transient Accumulation of Interfacial Charge in Photoconductor-Dielectric Systems

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This paper presents experimental results which reveal the occurrence of free-carrier accumulation (i.e., no trapping and no potential-energy barrier to injection) at a photoconductor-dielectric interface. The space-charge-limited current of the dielectric emerges as a key parameter in the process. Several predictions concerning the rate of accumulation of free charge at the interface and the dependence of that rate on applied voltage and current levels in the photoconductor are made using a qualitative model. The experiments were carried out on a model system, amorphous-Se—PVK [poly(*N*-vinyl carbazole)], where by means of light the Se is made a conductor of variable "dark" conductance. Direct observation of the development of interfacial charge has been possible by use of a double-relay technique. The variation with applied voltage and light intensity of the time for accumulation of a CV of charge conforms qualitatively with the predictions of the theoretical model.

I. INTRODUCTION

A recent paper[1] has described a limitation which can account for observed xerographic gain-vs-field curves in structures consisting of films of the polymer poly(*N*-vinyl carbazole) (PVK) overcoated with amorphous selenium. The limitation arises because even at relatively high fields the emission-limited current of the Se can be larger than the trap-free space-charge-limited currents (TFSCLC) in the PVK. In the experiments on the amorphous-Se–PVK system,[1] the assumption of capacitive field division throughout the measurement time for the emission-limited region was justified implicitly in terms of the high resistivities of the amorphous Se and PVK. By this criterion, structures incorporating a dielectric of relatively low resistivity on a high-resistivity dielectric, such as PVK or amorphous Se, would quickly develop essentially zero field in the more conducting dielectric. The experiments on the amorphous-Se–PVK system[1] indicated that the relative resistivities of the dielectrics in a composite structure are not, per se, the determining factor.

The purpose of this paper is to present experimental evidence which monitors the transition from

capacitive voltage division in a composite structure and which shows that this involves the transient accumulation of free charge at the interface between the two dielectrics. The basic feature of this paper is to stress that in the case of two dielectrics which possess no range limitation for carrier transport, no interfacial barriers to impede charge injection from one to the other, and in which no trapping occurs, that a limitation to current flow will still occur. This phenomenon is therefore different from those discussed in recent papers by Tabak and Scharfe[2] and Batra *et al.*[3] Tabak and Scharfe[2] discussed the limitation to current flow in a homogeneous photoreceptor (amorphous Se) due to the accumulation in the steady state of approximately 1 CV of trapped space charge in the sample bulk. This accumulation occurred after many transits in the amorphous Se. Batra *et al.*[3] analyzed theoretically the time dependence of current and voltage in a photoconductor-dielectric structure. In this case charge injection into the dielectric from the photoconductor was not allowed.

Section II of this paper contains a qualitative theoretical model. Sections III and IV describe the experimental technique and results, respectively. Section V will present a brief discussion of the results and their significance. The Appendix describes the effect of a field-dependent mobility, which occurs in PVK,[1] on the distribution of space charge for steady-state TFSCLC.

II. THEORY

Consider two dissimilar dielectrics in contact as indicated in Fig. 1. The layers have thicknesses d_1 and d_2 and dielectric constants K_1 and K_2. In order to calculate the voltage division it is tempting to postulate unique conductivities for the layers. The classic Maxwell-Wagner discussion of a layered dielectric structure does just this.[4] However, since the flow of current through an insulator is generally non-Ohmic, and may be controlled by space charge within the insulator or the nature of the contacts, the simple concept of "conductivity" does not lead to correct conclusions. Instead, one must specify the nonlinear dependence of current on voltage which applies with one particular choice of contacts. For example, amorphous Se is a good dielectric with a very long relaxation time as long as the only carriers in the material are thermally generated. However, because the hole mobility is relatively large, the maximum current which can be transported through a film of amorphous Se via injection is much greater than that due to the thermal generation of carriers.

To analyze the voltage division between the layers of the structure of Fig. 1, we define the following different voltages and true currents. The subscript

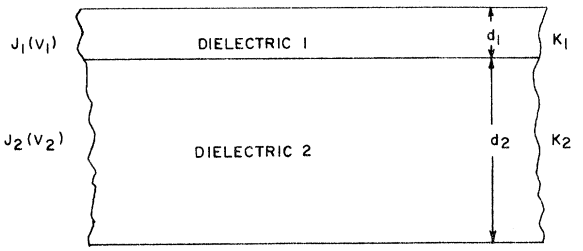

FIG. 1. Two dissimilar dielectrics in contact.

i is 1 or 2 for layer 1 or layer 2, respectively. $V_i(t)$ is the voltage across the layer at an arbitrary time, $V_i(0)$ is the initial voltage assuming capacitive division, and $V_i(\infty)$ is the final voltage for steady-state current. $J_i(V_i)$ is the current density in the isolated layer with neutral (noninjecting) contacts [a "conductivity" might be obtained from $J_i(V_i)$], $J_i'(V_i)$ is the current which flows when the two layers are in contact, and $J_{2\mathrm{SCL}}(V_2)$ is the space-charge-limited (SCL) current in layer 2 when in contact with layer 1. For simplicity it is assumed that no bulk trapping of charge occurs, and that there is no barrier to charge transport at the interface.

Consider conditions at the instant a voltage V_0 is applied across the structure. This initial voltage can be written $V_0 = V_1(0) + V_2(0)$, and $K_1 V_1(0)/d_1 = K_2 V_2(0)/d_2$. The voltages across the two dielectrics are therefore $V_1(0) = V_0 K_2 d_1/(K_1 d_2 + K_2 d_1)$ and $V_2(0) = V_0 K_1 d_2/(K_1 d_2 + K_2 d_1)$; i.e., the voltage divides capacitively. Upon application of V_0 a current $J_1'(V_1(0))$ will flow in layer 1 toward the interface and a current $J_2'(V_2(0))$ will flow in layer 2 away from the interface. In general these currents will not be equal and carriers will accumulate at, or be removed from the region near the interface. The voltages $V_1(t)$ and $V_2(t)$ will change as a result of this interfacial charge accumulation until $J_1' = J_2'$.

In order to determine $V_1(\infty)$ and $V_2(\infty)$ we must relate the currents $J_1'(V_1)$ and $J_2'(V_2)$ to $J_1(V_1)$ and $J_2(V_2)$, the latter having been measured witn the layers separated and with neutral contacts. We will suppose that the charge is carried by holes in both layers and that the bias V_0 makes layer 1 positive. Materials in which the carriers are electrons may be considered by letting V_0 have the opposite polarity. Then $J_1'(V_1) = J_1(V_1)$ since the density of carriers in layer 1 is unchanged by the presence of layer 2. If $J_2(V_2) > J_1'(V_1)$, charge will flow away from the interface in layer 2 until V_1 has increased and V_2 decreased sufficiently to make the currents equal. Even in the extreme case that $J_2'(V_2)$ is equal to $J_2(V_2)$ because of a negligible injection of carriers from layer 1, the final voltages will differ from the capacitive division values. On the other hand, if $J_1'(V_1(0)) > J_2(V_2(0))$, then the current

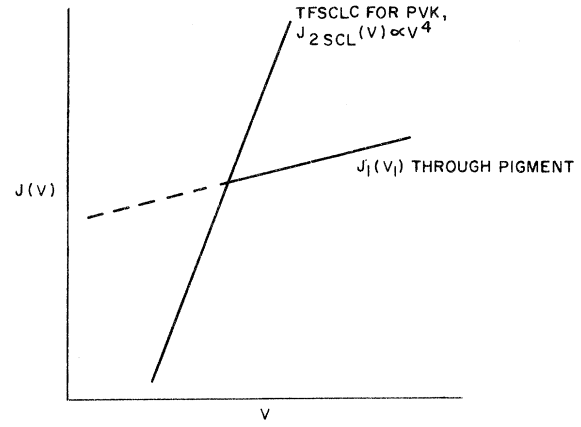

FIG. 2. Schematic plot of current density vs voltage for (i) TFSCLC in the active matrix PVK and (ii) dark current in hypothetical pigment.

$J_2'(V_2) = J_1'(V_1) + J_2(V_2)$, as long as $J_2'(V_2) \leq J_{2SCL}(V_2)$. We will assume that $J_2'(V_2) = J_1'(V_1)$ and that the charge flowing in layer 2 is just that injected from layer 1, since this is correct for the Se-PVK system. This assumption leads to the following two cases: (i) At the initial voltages $V_1(0)$ and $V_2(0)$, $J_1'(V_1(0)) < J_{2SCL}(V_2(0))$. In this case $J_2'(V_2(0)) = J_1'(V_1(0))$ and there is no charge accumulation at the interface. The fields within the layers remain constant at their initial values. (ii) At the initial voltages $V_1(0)$ and $V_2(0)$, $J_1'(V_1(0)) > J_{2SCL}(V_2(0))$. In this case charge will accumulate in the region of the interface until $J_1'(V_1(\infty)) = J_{2SCL}(V_2(\infty))$. Since the TFSCLC in PVK is much less than that in amorphous Se, it is apparent that a sensitizer which falls into case (i) when used with Se may fall into case (ii) when used with PVK. In case (ii) the photosensitivity will be substantially reduced because of the reduced voltage $V_1(\infty)$ in layer 1. However, when PVK is used the interesting possibility exists that, depending on the initial voltage V_0, charge accumulation may or may not occur. This is because for PVK, $J_{2SCL}(V_2) \propto V_2^4$ while the sensitizer is quite likely to exhibit a less extreme dependence of current on voltage. Then the situation shown in Fig. 2 occurs. At high voltage, $J_{2SCL}(V_2(0)) > J_1'(V_1(0))$ and no charge accumulates at the interface, but at lower voltage $J_{2SCL}(V_2(0)) < J_1'(V_1(0))$ and accumulation will occur.

A calculation of the time dependence of the accumulation of this transient interfacial charge, and its subsequent motion into the layer 2 as a space charge would be quite involved. Since we are concerned primarily in this article with the dependence of the interfacial-charge accumulation on the steady-state $J(V)$ characteristics of the layers and the total voltage V_0 applied to the composite structure, we make the following assumptions in order to estimate an approximate accumulation time. First, the development of space charge in layer 2 is neglected. In the steady state with a field-independent mobility one-third of the space charge lies next to the interface within one-ninth of the layer thickness. In the case of a field-dependent mobility a substantially larger fraction of the space charge lies close to the interface (see Appendix). It is therefore reasonable to neglect the motion of the interfacial charge into layer 2 in the Se-PVK system because of the strong field dependence of the mobility in PVK.[1] Furthermore, in this system the mobility of carriers in layer 1 (Se) is much greater than in layer 2 (PVK) so that charge injected into layer 1 arrives at the interface within a small fraction of the transit time of carriers through layer 2. Second, the diffusion of carriers, which would broaden the sharp charge sheet at the interface, is neglected. This is reasonable since diffusion broadens the charge sheet over a distance x such that $Ex \approx kT/e$. In this system E is $\gtrsim 10^4$ V cm^{-1}, and so x will be $\lesssim 2.5 \times 10^{-6}$ cm, or less than one one-hundredth of the thickness of the thinner layer (3×10^{-4} cm).

With these considerations and the fact that the initial displacement current which flows during establishment of capacitive division is made to occur in a very short time by the experimental arrangement, while the displacement current which flows during the accumulation of interfacial charge is included in the expression for $Q(t_Q)$ which follows, we may estimate the accumulation time from the equation for the accumulation of real charge:

$$Q(t) = \int_0^t [J_1'(V_1(t')) - J_2'(V_2(t'))]dt'$$

by setting $Q(t_Q) = C_2[V_1(0) - V_1(\infty)] \approx C_2 V_1(0)$ and approximating the integral by

$$Q(t_Q) = C_2 V_1(0) = t_Q [J_1'(V_1(0)) - J_{2SCL}(V_2(0))] .$$

C_2 in these expressions is the capacitance of layer 2 alone. This t_Q will give the approximate time to accumulate most of the charge; obviously the rate of accumulation will decrease as $J_1' \to J_2'$ and the approach to equilibrium will be asymptotic.

III. EXPERIMENTAL TECHNIQUE

In order to test the predictions of this model it was decided to use the amorphous-Se–PVK system. This offers several advantages. A detailed study of this system[1] has already indicated an absence of any potential-energy barrier to injection of holes from amorphous Se into PVK. Under most conditions there is no bulk trapping of holes either in the Se or PVK, or at the interface. The amorphous Se can be used as a simulated conducting dielectric of variable conductivity by illumination at different light levels. The light-generated emission-limited currents in amorphous Se can clearly be considered as equivalent to $J_1'(V_1)$ referred to in the above model. The variation of $J_1'(V_1)$ by varying the level

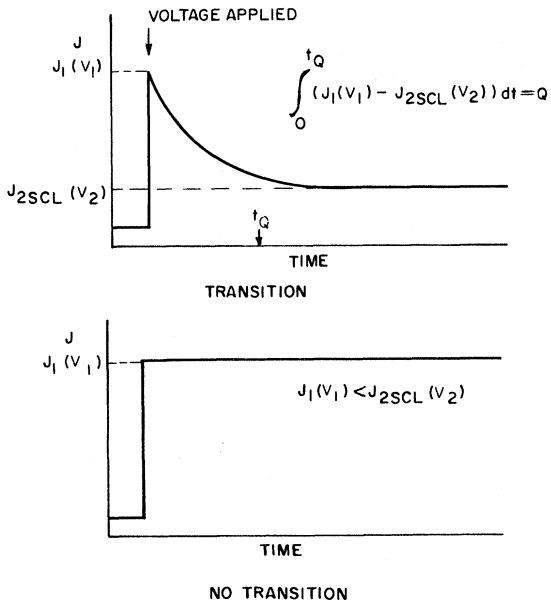

FIG. 3. Representation of current density versus time. In upper figure an accumulation of one CV of charge occurs at the interface in a time t_Q; in the lower figure no accumulation occurs.

of illumination is invaluable in testing the model, since it does not affect the value of $J_{2SCL}(V_2)$ of the PVK. Any attempt to vary $J_1'(V_1)$ of the conducting dielectric by changing the temperature would in addition greatly affect the value of $J_{2SCL}(V_2)$ which is thermally activated with an activation energy of 0.55 eV.[1] It is easily seen from the model that the transition from capacitive field division must involve a transient decay from one current value to another. This is indicated in Fig. 3. At the instant the voltage is applied to the system the field will divide capacitively and, under conditions of no bulk trapping and with any conceivable light level, the initial current will be an emission-limited current determined by the light level and the quantum efficiency for carrier generation is amorphous Se at that field.

If the emission-limited current $J_1'(V_1(0))$ is greater than $J_{2SCL}(V_2(0))$, the current through the composite structure will decay from $J_1'(V_1(0))$ to $J_{2SCL}(V_2(\infty))$ as the interfacial charge accumulates in a time t_Q which was estimated in Sec. III. In order to observe this transient decay in current a step-function voltage must be applied to the sample sufficiently fast that the field is established in a time much less than t_Q. The application of a fast-rising voltage pulse to insulating materials invariably produces a large capacitive current which will saturate any amplifier connected to measure the sample current. This leads to a dead time during which the observation of small-signal levels is impossible. The dead time can be substantially reduced by gating the input to the amplifiers during the establishment of the field. A double-relay method[5] was used; a schematic of the apparatus is given in Fig. 4. The mercury-wetted-contact relay 1 produces a voltage step which rises in a fraction of a microsecond with variable amplitude up to 900 V. Mercury-wetted-contact relay 2 shorts out the load resistor R_L during the establishment of the voltage level. The coils of these two relays are connected in parallel and are energized for a single operation by closing the microswitch. As the speed of response of a relay depends largely on the energizing current, the variable resistor R_D is used to delay the opening of relay 2 by a time Δt with respect to the closing of relay 1. The limiting factor in this method is the reproducibility of individual relay operations. The useful minimum time of Δt is about 50 μsec. The oscilloscope is triggered on the closing of relay 1. This provides a common origin for the time scale and successive sweeps will superimpose. It is essential, of course, that the value of R_L is sufficiently small to make the time constant determined by the specimen capacity and the load resistor much smaller than the time constant to be measured. By keeping the microswitch closed, the approach to steady-state conditions could be observed and measurements made in this regime. Illumination was achieved using a dc 100-W tungsten-iodine lamp using narrow-pass-band interference filters. It was found essential to use heat-absorbing filters to prevent any heating of the sample. Unfiltered steady illumination heated the sample and thus caused J_{2SCL} to change with time. The samples were identical to those used in earlier studies of the response of the amorphous-Se-PVK system to intense short light pulses.[1] 10 μ of PVK were deposited from a solution of PVK in toluene and cyclohexanone on NESA glass and overlayed with 3 μ of amorphous Se which was provided with a semitransparent Au electrode.

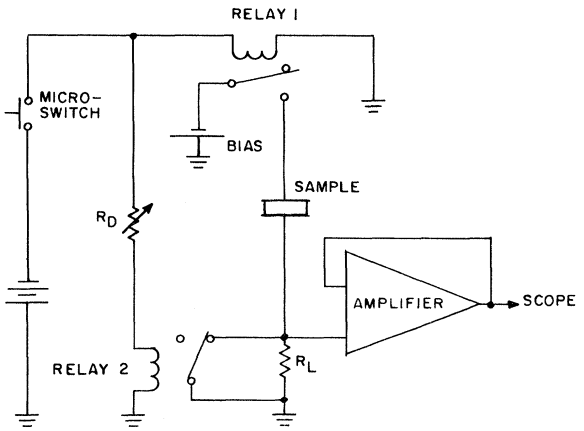

FIG. 4. Schematic of double-relay apparatus.

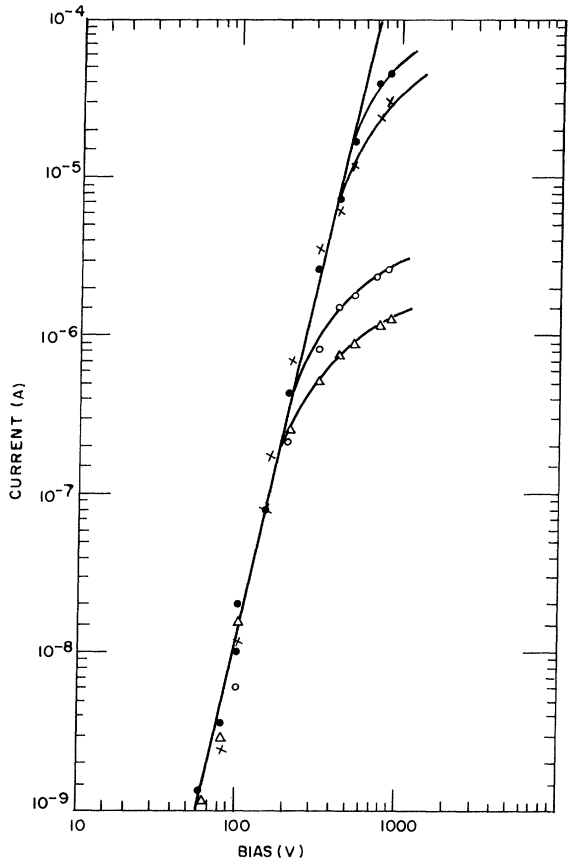

FIG. 5. Steady-state current density versus applied voltage for 3-μ amorphous-Se layer on 10 μ of PVK. The different symbols correspond to different light levels and fall on a common curve at low voltage. The data deviate from this common curve at higher voltages with increasing light intensity.

IV. EXPERIMENTAL RESULTS

Figure 5 shows a plot of the steady-state current through the Se-PVK structure as a function of applied voltage and at different light intensities. The Au electrode was biased positive; the light was incident through this electrode and strongly absorbed in the Se. The curve is explained in the following way. At low voltages ($\lesssim$ 200 V) the current through the composite structure is independent of light intensity and is approximately proportional to V^4. At about 250 V for the lowest light intensity, the current changes its voltage dependence to that of emission-limited currents in Se (approximately proportional to $V^{0.5}$). At higher light intensities a similar change occurs at increasingly higher voltage. This experimentally verifies the transition from the TFSCLC in PVK to the emission-limited current in amorphous Se. The transition occurs at a voltage when the emission current can no longer supply the space-charge current demanded by the PVK. Clearly the transition voltage will increase as the light-dependent emission current increases with higher light levels.

In Fig. 6 two of the experimental curves of Fig. 5 have been redrawn in a schematic form in order to explain the transient responses of the Se-PVK system to the step-function voltage. Immediately we draw a parallel between this plot and that in Fig. 2 on which the model is based. The emission-limited curves are extrapolated to low fields and represented by the dashed lines. The transition from $J_1'(V_1)$ to $J_{2SCL}(V_2)$ is denoted by the arrows corresponding to the transition from capacitive division. For ease of presentation different regimes will be discussed and predictions of the expected behavior made. These will then be compared with experimental observation.

A. Constant Light Intensity—Variable-Step Bias

At low bias $J_1'(V_1) - J_{2SCL}(V_2)$ is very large. We have estimated the decay time constant t_Q by $C_2V_1 = t_Q[J_1'(V_1(0)) - J_{2SCL}(V_2(0))]$. t_Q is approximately the time required for the initial surface charge Q to accumulate at the interface. If t_Q is very much less than the transit time T_R of holes in PVK at

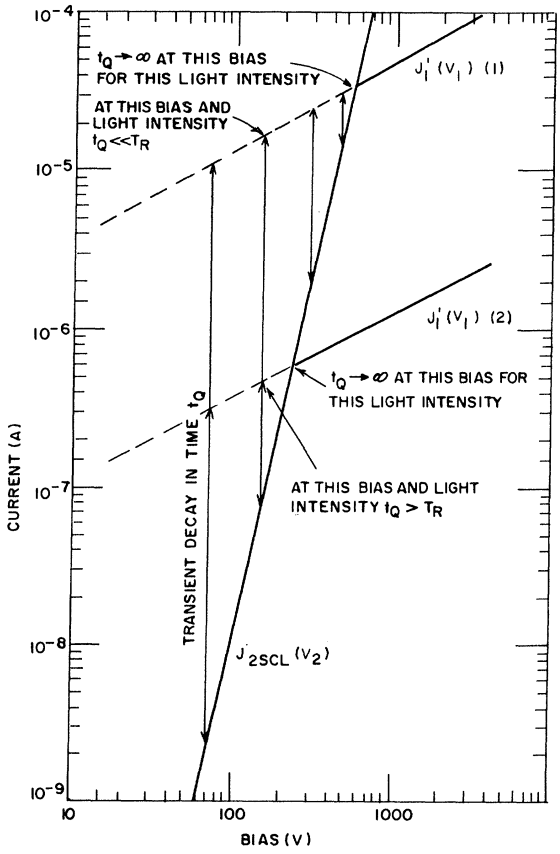

FIG. 6. Schematic form of two of the experimental curves shown in Fig. 5.

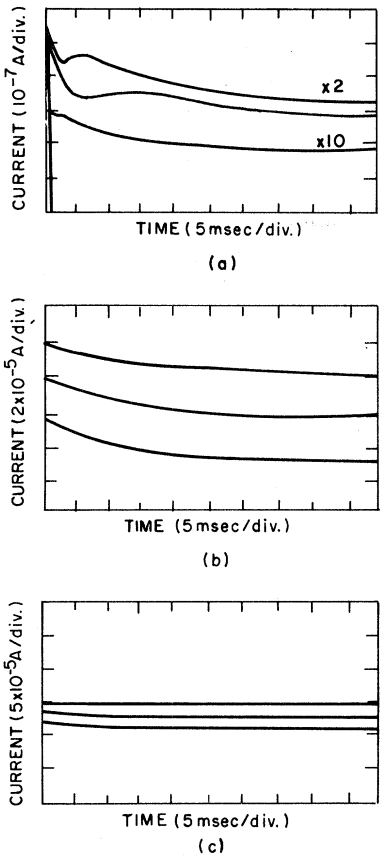

FIG. 7. Transient response to step-function voltage at the same light intensity as in curve 1, Fig. 6. (a) At low voltage the response is determined by the transit of holes through the PVK. The lower curve corresponds to a bias of 155 V, the middle to 84 V, and the upper to 105 V. (b) At intermediate voltages the response is determined by the rate of accumulation of one CV of charge at the interface. From the lowest trace upwards the biases are 400, 500, and 600 V. (c) At the highest voltages no accumulation of charge occurs at the interface and no current decay occurs. From the lowest trace upwards the biases are 700, 800, and 900 V.

this bias, then, as far as subsequent motion through the PVK is concerned, the total charge arrives at the interface instantaneously. We therefore, expect to observe the time-resolved transit of holes through the PVK. As the bias is increased the transit time of these holes will decrease in the usual way (proportional to $V^{-2.3}$, see Ref. 1). In addition, however, $J_1'(V_1(0)) - J_{2SCL}(V_2(0))$ becomes smaller faster than Q increases, and at some voltage T_R and t_Q will be equal, while at higher biases T_R will be less than t_Q. In this region where $T_R < t_Q$ the transient will trace out the buildup of charge at the interface and the transit of carriers, through the PVK will not be resolvable. As the bias is increased still further t_Q will become larger until, at the point of intersection of $J_1'(V_1(0))$ and

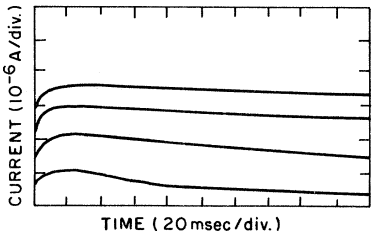

FIG. 8. Transient current response taken at lower light intensity (curve 2 in Fig. 6) than in Fig. 7. From the lowest trace upwards the biases are 150, 230, 305, and 380 V.

$J_{2SCL}(V_2(0))$, where the difference between these currents is zero, $t_Q = \infty$. At this point and beyond, charge is not accumulated at the interface since the PVK can always carry the full emission currents available at this light intensity. Figures 7(a)–7(c) show the transient response taken at constant light intensity, corresponding to curve 1 in Fig. 6, as a function of bias. Figure 7(a) taken at relatively low bias where t_Q is $< T_R$ shows the transit of holes through the PVK. In Figs. 7(b) and 7(c) the decay is already dominated by an increasing t_Q and no transit is observable. The time t_Q goes to ∞ at ~ 900 V which is close to the estimated bias at which $J_1'(V_1(0))$ and $J_{2SCL}(V_2(0))$ intersect at this light level.

It is predicted from Fig. 6 that at the lower light level, curve 2, the voltage at which $t_Q \to \infty$ should occur at about 300 V instead of 900 V for the higher light level. In addition, since for a given bias the difference $J_1'(V_1(0)) - J_{2SCL}(V_2(0))$ is smaller for the lower light level, for the same bias the time t_Q should be longer for the smaller light level than the higher one. This prediction is completely verified by the experimental curves in Fig. 8.

B. Constant-Step Bias—Variable Light Intensity

A rather unusual observation was made in this regime. This observation, which at first was rather mystifying, is in fact quite predictable in terms of the model. Consider a bias of 150 V. The J-t traces in Fig. 7(a) show quite clearly the

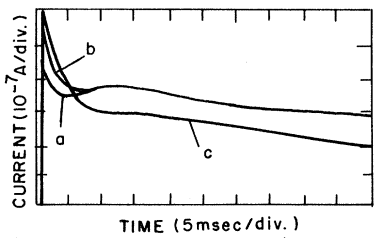

FIG. 9. Transient current response taken at constant bias at different light intensities: curve (a), full intensity; curve (b), $\frac{1}{3}$ intensity; curve (c), $\frac{1}{10}$ intensity.

hole transit through the PVK. However, in Fig. 8, made at this same bias but lower light intensity no hole transit is observable and the current decay is dominated by a much longer time constant. Returning to Fig. 6 the explanation is obvious. At a given bias the reduction in the light level leads to a decrease in $J_1'(V_1(0)) - J_{2SCL}(V_2(0))$ and an increase in t_Q such that, in this case, t_Q changes from being less than T_R to being greater. The evolution of this transition is shown in Fig. 9 where, at a fixed bias of 84 V, a transit is observed at full light intensity but becomes barely observable when the light is attenuated by a factory of 10.

V. SUMMARY

The model proposed to establish a physical basis for understanding the transition from capacitive division has been verified. It is shown that even if the dependence of current on electric field were linear for the separate dielectrics (pigment and transport medium), the criterion as to whether interfacial-charge accumulation occurs (which results in "resistive" field division) is not determined by "conductivities," i. e., by the product of the thermal equilibrium density of holes and their mobility, but by the maximum (space-charge-limited) current in the second (transport) layer, as compared with the dark current in the pigment.

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APPENDIX: DISTRIBUTION OF SPACE CHARGE

In the steady state we solve Poisson's equation

$$-\frac{\partial E}{\partial x} = \frac{\partial^2 \varphi}{\partial x^2} = -4\pi \frac{\rho(x)}{\epsilon} \tag{1}$$

subject to current continuity, J = constant, and neglecting diffusion

$$J = \rho(x)\mu E(x)\,, \tag{2}$$

with the boundary conditions

$$\left.\begin{aligned} E &= 0 \\ \varphi &= 0 \end{aligned}\right\} \text{ at } x = 0\,,$$
$$\varphi = V \quad \text{at } x = a\,.$$

Then with constant mobility μ

$$\int_0^x \rho(x)\,dx = Q(x) = \frac{3}{2}\frac{\epsilon}{4\pi a} V\left(\frac{x}{a}\right)^{1/2}, \tag{3}$$

$$Q(a) = \tfrac{3}{2} CV\,, \tag{4}$$

where C is the capacitance of the layer. These results are well known.[6] If now

$$\mu = \mu_0(x)[E(x)/E_0]^n\,, \tag{5}$$

one finds that

$$Q(x) = \frac{n+3}{n+2}\frac{\epsilon}{4\pi a} V\left(\frac{x}{a}\right)^{1/(n+2)}. \tag{6}$$

With $n = 2$, as is appropriate for PVK,[1] Eq. (6) becomes

$$Q(x) = \frac{5}{4}\frac{\epsilon}{4\pi a} V\left(\frac{x}{a}\right)^{1/4}$$

and thus for $x = 0.01a$, $Q(x) = 0.317Q(a)$.

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