Phototransport under the presence of a small steady-state photocarrier grating

Yuan-Min Li*
Gordon McKay Laboratory, Harvard University, Cambridge, Massachusetts 02138
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By taking into account space-charge effects, *general* expressions for carrier distributions and photocurrent are derived for the case of a small steady-state photocarrier grating (SSPG) in a finite electric field. Conditions are specified under which the continuity equations can be linearized and solved analytically. It is shown that the approach of spatially averaging local resistivity in the previous treatments of SSPG transport can be flawed. It is argued that the drift motion of electrons and holes is predominantly bipolar rather than ambipolar as asserted earlier. The prerequisites for determining the ambipolar diffusion length L by the SSPG technique are identified and compared to those obtained before. It is demonstrated that under the weak-field condition, L can be evaluated *without* the lifetime-regime or ambipolarity restriction, particularly when the recombination lifetime τ is known. When the dielectric relaxation time is far shorter than τ, the transport equation is reduced to the familiar formula derived by assuming space-charge neutrality. Also illustrated is the possibility of deducing the ratio of electron-to-hole drift mobility through the electric field dependence of photocurrent under the strong-field condition.

I. INTRODUCTION

The ambipolar diffusion length L is largely a measure of the diffusion length of the less mobile type of carriers (presumably holes) and is an important parameter for bipolar semiconductor devices such as transistors and solar cells.[1-8] Recently, the steady-state photocarrier grating (SSPG) technique was proposed to measure L in those semiconductors such as hydrogenated amorphous silicon (a-Si:H) and its alloys, in which L is on the order of a fraction of 1 μm.[1-3] For these materials the SSPG technique is preferable over the more traditional surface photovoltage technique[9] which tends to overestimate L due to carrier drift in the built-in electric field near the junction region.[3] In the original analysis of SSPG, a simple formula relating photoconductivity to L was obtained under the assumption of space-charge neutrality.[1] Subsequently, by numerically solving the steady-state continuity equations for photoelectrons and photoholes, it was recognized that the simple formula is valid only under the conditions of fast dielectric relaxation and weak electric field (see discussion below).[2] More recently, an expression for steady-state photoconductance under the presence of a small photocarrier grating was obtained for the case of a negligibly small, externally applied electric field.[4] This paper, which is based on a conference report,[10] presents general formulas for the distributions and transport of photocarriers in a finite electric field by allowing for space-charge non-neutrality and electric-field grating. In Sec. II, conditions are stated under which the continuity equations can be made linear. These equations are then solved analytically to express the gratings of photoelectrons, photoholes, and the electric field in terms of several physically meaningful ratios. Section III presents an analysis of transport in SSPG using the continuity equation for electric charge. It is shown that the earlier approach for SSPG transport by spatially averaging the local conductivity[1-4] is inadequate unless the ratio of photocarrier lifetime τ to dielectric relaxation time $\tau_{\rm rel}$ is sufficiently high. It will become evident that electron and hole transport is bipolar rather than ambipolar as previously believed.[5] Section IV discusses the application of the transport formula to the study of the transport of the less mobile type of carriers (holes). The conditions under which the general photocurrent formula[10] can be reduced to the popular, simple formula[1,3] will be specified and compared to those proposed before.[2,4] It is suggested that, in principle, L can be determined even if the lifetime regime[2] or the ambipolarity[4] requirement ($\tau/\tau_{\rm rel} \gg 1$) is dissatisfied. This becomes particularly feasible once τ is known.[10] Also discussed is the possibility of deducing the ratio of electron to hole drift mobility (μ_n/μ_p) by studying the field dependence of photocurrent when diffusion is negligible compared to drift.

II. PHOTOCARRIER AND ELECTRIC-FIELD GRATINGS

The original idea[1] of the SSPG technique is to observe the effect on photoconductivity of a small spatial modulation (or grating) of photocarrier concentration whose amplitude depends *inter alia* on L. Photocarrier grating is produced by an optical excitation grating from two interfering laser beams I_1 and I_2, which satisfy $I_1 \gg I_2$. This leads to a small grating of optical excitation riding on a much stronger uniform background generation of electron-hole pairs. For penetrating illumination, the generation rate $G(x)$ is given by[1]

$$G(x)=G_0+\Delta G(x)=\kappa[I_1+I_2+2\gamma_0\sqrt{I_1 I_2}\cos(kx)] , \tag{1}$$

where κ stands for the optical absorption coefficient at the laser wavelength λ, $k=2\pi/\Lambda$, where

$\Lambda=\lambda/[2\sin(\delta/2)]$ is the optical grating period, δ is the angle between the two beams, γ_0 $(0<\gamma_0\leq 1)$ is the interference-quality factor,[1] x is the coordinate perpendicular to the interference fringes, and $G_0=\kappa(I_1+I_2)$ is the generation rate from the uniform background illumination. The excitation grating

$$\Delta G(x)=2\kappa\gamma_0\sqrt{I_1 I_2}\cos(kx)=G_1\cos(kx) \tag{2}$$

has an amplitude of $G_1=2\kappa\gamma_0(I_1 I_2)^{1/2}$. The densities of electrons and holes, $N(x)$ and $P(x)$, are modulated by ΔG to become

$$N(x)=N_0+\Delta N(x)=N_0+\Delta N \tag{3}$$

and

$$P(x)=P_0+\Delta P(x)=P_0+\Delta P\ , \tag{4}$$

where N_0 and P_0 are the uniform background carrier densities in the absence of coherence between I_1 and I_2, ΔN and ΔP are density gratings of photoelectrons and photoholes. It should be emphasized that N and P here represent the *total* densities of electrons and holes, rather than the mobile carriers alone (as is the case in Ref. 4). For noncrystalline semiconductors (e.g., *a*-Si:H) with localized band-tail states,[11] N (P) is the sum of free *and* trapped electrons (holes). For undoped samples or doped samples with low dark current relative to photocurrent, $N_0=P_0$ as a result of the prevailing local charge neutrality in the bulk (assuming that Ohmic contacts[12,13] are used and that the current is not space-charge limited[14]). Since $G_1\ll G_0$, ΔN and ΔP are perturbations to N_0 and P_0, respectively. Thus, the following discussion regards as approximately constant the local drift mobility[15] and diffusion coefficient for electrons (holes), μ_n (μ_p) and D_n (D_p), which generally vary with carrier densities.[2,13–15] In the present context, μ and D refer to the averaged mobility and diffusion coefficient over all mobile *and* trapped carriers. In the unlikely event that the average mobility of photocarriers differs significantly from that of dark carriers, the subsequent analysis is applicable only to the situation when the photocurrent is considerably larger than the dark current.

Let J_n and J_p be, respectively, the fluxes of electrons and holes in the x direction. J_n and J_p are made up of diffusion and drift terms:

$$J_n=-D_n\frac{\partial N}{\partial x}-N\mu_n E\ , \tag{5}$$

$$J_p=-D_p\frac{\partial P}{\partial x}+P\mu_p E\ , \tag{6}$$

where E is the electric field,

$$E=E(x)=E_0+\Delta E(x)\ , \tag{7}$$

consisting of an externally applied field E_0 and a field grating $\Delta E(x)$. The field grating is brought about by unequal diffusion rates and/or opposite drifts of electrons and holes. ΔE is related to the space-charge density $e(P-N)=e(\Delta P-\Delta N)$ by the Poisson equation (in rationalized units)

$$\frac{dE}{dx}=\frac{d(\Delta E)}{dx}=\frac{e}{\epsilon\epsilon_0}(\Delta P-\Delta N)\ , \tag{8}$$

where ϵ is the dielectric constant of the sample, ϵ_0 the dielectric permittivity of the vacuum, and e the electronic charge. It will become evident shortly that ΔE, and therefore space charge, is nonvanishing unless $D_n=D_p$ and $E_0=0$ (in which case there is no electric current). In writing Eqs. (5) and (6), it is assumed that if there is a high density of dark carriers, these have the same drift mobility as that of the photocarriers. This assumption seems reasonable if the majority of the traps (e.g., in noncrystalline materials[11]) are not much deeper than the thermal energy $k_B T$ (where k_B is the Boltzmann constant and T the temperature), such that excess carriers in the extended states are in near equilibrium with those in the localized states.

Now let R be the *recombination* (not deep trapping) rate of excess electrons and holes. $R=R_0+\Delta R$, where $R_0=G_0$ is the recombination rate in the absence of any photocarrier grating ($\Delta G=0$), and ΔR results from the variations of local densities of photocarriers. Note that ΔR is the same for photoelectrons and photoholes, since recombination takes place between the two types of carriers. The *steady-state* continuity equations for ΔN and ΔP are[6,7]

$$D_n\frac{d^2(\Delta N)}{dx^2}+\mu_n E\frac{d(\Delta N)}{dx}+\mu_n N\frac{dE}{dx}-\Delta R+\Delta G=0, \tag{9}$$

$$D_p\frac{d^2(\Delta P)}{dx^2}-\mu_p E\frac{d(\Delta P)}{dx}-\mu_p P\frac{dE}{dx}-\Delta R+\Delta G=0\ . \tag{10}$$

For small photocarrier gratings, ΔR can be approximated by

$$\Delta R=\frac{1}{2\tau}(\Delta N+\Delta P)\ , \tag{11}$$

where τ is the photocarrier recombination lifetime, or the small-signal photocurrent response time from the steady state,[2,13–16] under a uniform excitation of G_0:

$$\frac{1}{\tau}=\left.\frac{\partial R}{\partial N}\right|_{G_0}=\left.\frac{\partial R}{\partial P}\right|_{G_0}\ , \tag{12}$$

where the derivatives are to be taken with respect to the density of photocarriers (excluding dark carriers). Note that in noncrystalline semiconductors of a high density of traps τ can be much greater than the free-carrier lifetime, as most of the carriers are immobile.[13–16] Combining Eqs. (2), (8), and (11) with Eqs. (9) and (10), one gets the coupled equations for ΔN and ΔP:

$$D_n\frac{d^2(\Delta N)}{dx^2}+\mu_n E\frac{d(\Delta N)}{dx}+\frac{e}{\epsilon\epsilon_0}\mu_n N(\Delta P-\Delta N)-\frac{1}{2\tau}(\Delta N+\Delta P)+G_1\cos(kx)=0 \tag{13}$$

and

$$D_p\frac{d^2(\Delta P)}{dx^2}-\mu_p E\frac{d(\Delta P)}{dx}-\frac{e}{\epsilon\epsilon_0}\mu_p P(\Delta P-\Delta N)$$
$$-\frac{1}{2\tau}(\Delta N+\Delta P)+G_1\cos(kx)=0\ . \quad (14)$$

The exact solutions to Eqs. (13) and (14) are prohibitively complicated because of the nonlinear terms such as $\Delta E\, d(\Delta N)/dx$ and $\Delta N\,\Delta P$. The problem is enormously simplified when the above equations are *linearized* by replacing E, N, and P with E_0, N_0, and P_0, respectively. Then one gets sinusoidal solutions of the following form:

$$\Delta N=\overline{\Delta N}\cos(kx+\phi_n)=N_1\cos(kx)+N_2\sin(kx)\ , \quad (15)$$

$$\Delta P=\overline{\Delta P}\cos(kx+\phi_p)=P_1\cos(kx)+P_2\sin(kx)\ , \quad (16)$$

$$\Delta E=\overline{\Delta E}\cos(kx+\phi_E)=E_1\cos(kx)+E_2\sin(kx)\ , \quad (17)$$

where the ϕ's are the phases with respect to ΔG. The linearization approximation will be justified if the oscillation *amplitudes* of the nonlinear terms are much smaller than those of the linear terms (see a similar discussion in Ref. 4). When E_0 is low, such that carrier distribution is largely controlled by diffusion rather than drift in an external field, the linearization requirement is

$$D_p k\gg\mu_p\,\overline{\Delta E}\ , \quad (18)$$

because the amplitudes for $D_p\,d^2(\Delta P)/dx^2$ and $\mu_p\Delta E\,d(\Delta P)/dx$ [and $\mu_p\,\Delta P\,d(\Delta E)/dx$] in Eq. (10) are $D_p\,\overline{\Delta P}\,k^2$ and $\mu_p\,\overline{\Delta E\,\Delta P}\,k$, respectively. A similar relation

$$D_n k\gg\mu_n\,\overline{\Delta E} \quad (19)$$

can be obtained from Eq. (9). Note the well-known Einstein relation[6]

$$\mu_p=\frac{eD_p}{k_B T} \quad (20)$$

and

$$\mu_n=\frac{eD_n}{k_B T} \quad (21)$$

for electrons and holes; Eqs. (18) and (19) become roughly

$$k_B T>e\,\overline{\Delta E}\,\Lambda\ , \quad (22)$$

i.e., the thermal energy must be greater than the maximum potential energy associated with the oscillating internal electric field of amplitude $\overline{\Delta E}$ and period Λ. Obviously, this precondition can be satisfied if G_1, and hence the photocarrier grating, is sufficiently small. Note that Eq. (22) is analogous to, and more transparent than, the condition proposed in Ref. 4 [Eq. (3.12)]. When E_0 is strong such that carrier drift is significant in determining the photocarrier gratings (the weak-field and strong-field conditions will be spelled out in Sec. IV), an additional requirement for the linearization of Eqs. (9) and (10), or Eqs. (13) and (14), arises,

$$E_0\gg\overline{\Delta E}\ , \quad (23)$$

which is derived by comparing the amplitudes of $\mu_p E_0\,d(\Delta P)/dx$ and $\mu_p\,\Delta E\,d(\Delta P)/dx$ [or $\mu_p\,\Delta P\,d(\Delta E)/dx$] in Eq. (10). The self-consistency of the linearization of Eqs. (13) and (14) can be checked *a posteriori* using Eq. (22) and/or Eq. (23) by observing

$$\overline{\Delta E}=[(E_1)^2+(E_2)^2]^{1/2} \quad (24)$$

and using the expressions for E_1 and E_2 to be given below.

Several parameters need to be introduced now. The ambipolar diffusion length L is defined as

$$L=\sqrt{\tau D}\ , \quad (25)$$

where D is the ambipolar diffusion coefficient given by[6,7] [note $N_0=P_0$ and Eqs. (20) and (21)]

$$D=\frac{\mu_n D_p+\mu_p D_n}{\mu_n+\mu_p}=\frac{2D_n D_p}{D_n+D_p}\ . \quad (26)$$

The dielectric relaxation time $\tau_{\rm rel}$ of the sample under photogeneration G_0 is related to the conductivity σ by[7,13]

$$\tau_{\rm rel}=\frac{\epsilon\epsilon_0}{\sigma}=\frac{\epsilon\epsilon_0}{eN_0(\mu_n+\mu_p)} \quad (27)$$

or $\tau_{\rm rel}=8.8\times10^{-14}\epsilon/\sigma$ (in sec) when σ is expressed in units of $(\Omega\,{\rm cm})^{-1}$.

It is straightforward to solve a set of four linear algebraic equations for N_1, N_2, P_1, and P_2 obtained by inserting Eqs. (15) and (16) into the linearized Eqs. (13) and (14). After lengthy manipulations and rearrangements of various terms (the tedious bookkeeping details will not be reproduced here for sake of space), with repeated use of Eqs. (20) and (21), one gets

$$N_1=\frac{G_1\tau}{W}\left\{\left[\left[a+\frac{(b+1)^2}{4b}l^2\right](1+l^2)+d^2\right]\times\left[\frac{1+b}{2b}l^2+a\right]+\frac{1-b}{2b}d^2\right\}\ , \quad (28)$$

$$P_1=\frac{G_1\tau}{W}\left\{\left[\left[a+\frac{(b+1)^2)}{4b}l^2\right](1+l^2)+d^2\right]\times\left[\frac{1+b}{2}l^2+a\right]+\frac{b-1}{2}d^2\right\}\ , \quad (29)$$

$$N_2=-\frac{G_1\tau d}{W\sqrt{b}}\left[\left[a+\frac{(b+1)^2}{4b}l^2\right](1+l^2)+d^2+\frac{b^2-1}{4b}l^2+\frac{b-1}{2}a\right]\ , \quad (30)$$

$$P_2=\frac{G_1\tau d\sqrt{b}}{W}\left[\left[a+\frac{(b+1)^2}{4b}l^2\right](1+l^2)+d^2+\frac{1-b^2}{4b}l^2+\frac{1-b}{2b}a\right]\ , \quad (31)$$

where

$$W \equiv \left[\left[a + \frac{(b+1)^2}{4b} l^2 \right] (1+l^2) + d^2 \right]^2 + \frac{(b-1)^2}{4b} d^2 , \tag{32}$$

and where l, a, b, and d are given by

$$l \equiv kL = \frac{2\pi L}{\Lambda} , \tag{33}$$

$$a \equiv \frac{\tau}{\tau_{\text{rel}}} , \tag{34}$$

$$b \equiv \frac{\mu_n}{\mu_p} = \frac{D_n}{D_p} , \tag{35}$$

and

$$d \equiv k \sqrt{\mu_n \mu_p} \tau E_0 = \frac{2\pi \sqrt{\mu_n \mu_p} \tau E_0}{\Lambda} . \tag{36}$$

The meanings of the dimensionless ratios l, a, b, and d are self-evident. The likelihood of blurring the photocarrier gratings by diffusion depends on l. The degree of blurring the photocarrier gratings by drift in an external electric field is gauged by d, the ratio of the geometric mean of the electron and hole drift length to Λ. The photocarrier gratings are most pronounced when l and d are small. When l and/or d are large, the photocarrier gratings are smeared out by diffusion and/or drift (ΔN and ΔP vanish). The tendency of space-charge generation due to different diffusion coefficients of electrons and holes is reflected in b: The bigger the deviation of b from unity, the stronger the tendency. The material's ability to neutralize photocarrier-related space charge is measured by a, since fast dielectric relaxation (relative to recombination) prevents the buildup of local charge and field grating. Note that due to the symmetry of the continuity equations, the solution for ΔP is obtained by substituting d and b with $-d$ and $1/b$, respectively, in the expression for ΔN. The totality of a, b, l, and d determines the amplitude and phase of the photocarrier gratings. As expected, N_2 and P_2, which are the "out-of-phase" components of ΔN and ΔP (with respect to ΔG), are induced by the external field and diminish when $E_0 = 0$ ($d = 0$). Space-charge neutrality ($\Delta N = \Delta P$) is well maintained only if either a is very large (compared to bl^2) or b is unity, and only for small E_0 (d) (see Sec. IV).

The electric-field grating can be computed according to Eq. (8) using Eqs. (15)–(17) and (28)–(31). One thereby obtains

$$E_1 = -E_0 \left(\frac{G_1 \tau}{N_0} \right) \frac{a}{W} \left[\left[a + \frac{(b+1)^2}{4b} l^2 \right] (1+l^2) + d^2 - \frac{(b-1)^2}{4b} l^2 \right] \tag{37}$$

and

$$E_2 = \frac{e G_1 \tau}{\epsilon \epsilon_0 k W} \frac{b^2-1}{2b} \left[\left[a + \frac{(b+1)^2}{4b} l^2 \right] (1+l^2) l^2 + d^2 \right] . \tag{38}$$

It is evident that the "in-phase" component of $\Delta E (E_1)$ is caused by the external field E_0 while the "out-of-phase" component of ΔE (E_2) is mainly related to the difference in electron and hole diffusion coefficients signified by b ($\Delta E_2 = 0$ if $b = 1$). The physical origin of ΔE in the case of zero E_0 is the same as the Dember effect.[6,7] By inserting some trial values for various parameters into Eqs. (37) and (38) (e.g., b near 10, l and d near unity), one can verify that the linearization conditions (22) and (23) are satisfied if a is close to or greater than unity and $G_1 \tau < 1 \times 10^{20}$ cm^{-3} ($\ll N_0$). If a is far smaller than unity, however, the space-charge effect can be severe (with a significant internal field grating) for a finite $G_1 \tau$ when neither l nor d is much greater than unity. In this case, not to be further examined, the above solutions for photocarrier and field gratings become invalid, and photocarrier diffusion and drift in *internal* as well as external field are closely coupled.

III. PHOTOTRANSPORT

In the previous discussions on phototransport in the presence of a small SSPG,[1–5] the photoconductivity was computed, without proof of validity, by averaging the local resistivity or inverse conductivity (in a "series resistor" model) according to

$$\sigma_{\text{ave}} = \left[\frac{1}{\Lambda} \int_0^{\Lambda} \frac{dx}{\sigma(x)} \right]^{-1} , \tag{39}$$

where

$$\sigma(x) = \sigma_0 + e[\mu_n \Delta N(x) + \mu_p \Delta P(x)] \tag{40}$$

and where $\sigma_0 = e(N_0 \mu_n + P_0 \mu_p) = eN_0(\mu_n + \mu_p)$. It will be shown in the analysis to follow that, unless a is much greater than l^2 [$\tau/\tau_{\text{rel}} \gg (2\pi L/\Lambda)^2$], this recipe of relating σ_{ave} to the measured photocurrent is incorrect.

The current density j will be written as $j(I_1, I_2)$ and $j(I_1 + I_2) = j_0$, respectively, for the two cases when I_1 and I_2 do and do not interfere with each other. The continuity equation for electric charge is

$$\frac{\partial Q}{\partial t} + \frac{\partial j}{\partial x} = 0 , \tag{41}$$

where Q is the charge density. In the *steady state*, $\partial Q/\partial t = 0$; consequently $j(I_1, I_2)$ must be constant at all x. This occurs because the spatially varying diffusion-related current [$D_n \, d(\Delta N)/dx \neq D_p \, d(\Delta P)/dx$] is locally offset by a spatially varying component of the drift current of equal magnitude and opposite direction. From Eqs. (5) and (6), one has

$$j(I_1, I_2) = e(J_p - J_n) = e \left[D_n \frac{dN}{dx} - D_p \frac{dP}{dx} + E(\mu_n N + \mu_p P) \right] = \text{const} , \tag{42}$$

where N, P, and E are given by Eqs. (3), (4), and (7). In view of the fact that ΔN, ΔP, and ΔE are sinusoidal func-

tions given by Eqs. (15)–(17), the average (or integration) on both sides of Eq. (42) over a period Λ eliminates those terms *linear* in ΔN, ΔP, and ΔE to yield[10]

$$j(I_1,I_2)=j_0+\frac{e}{\Lambda}\int_0^{\Lambda}\Delta E(\mu_n\,\Delta N+\mu_p\,\Delta P)dx$$
$$=j_0+\Delta j\ , \tag{43}$$

where

$$j_0\equiv j(I_1+I_2)=e(\mu_n N_0+\mu_p P_0)E_0$$
$$=eN_0(\mu_n+\mu_p)E_0 \tag{44}$$

is the background current density (when $\Delta G=0$). The change in current density resulting from an illumination grating is governed by the photocarrier *and* electric field gratings

$$\Delta j\equiv j(I_1,I_2)-j_0=\frac{e}{\Lambda}\int_0^{\Lambda}\Delta E(\mu_n\,\Delta N+\mu_p\,\Delta P)dx\ . \tag{45}$$

Equation (45) shows that, strictly speaking, the field grating is never negligible (and space charge must exist) for nonvanishing Δj. The second-order terms are critical for *transport* analysis. From Eqs. (8) and (15)–(17), the field grating can be written as

$$\Delta E=\frac{e}{\epsilon\epsilon_0 k}[(N_2-P_2)\cos(kx)+(P_1-N_1)\sin(kx)]\ . \tag{46}$$

Inserting Eqs. (15), (16), and (46) into Eq. (45), one finds

$$\Delta j=\frac{e^2}{2\epsilon\epsilon_0 k}(\mu_n+\mu_p)(N_2P_1-N_1P_2)\ . \tag{47}$$

Inserting Eqs. (28)–(31) into Eq. (47), one finally arrives at the *general* transport formula,[10]

$$\Delta j=-\frac{j_0}{2}\left[\frac{G_1\tau}{N_0}\right]^2\frac{a(a+l^2)}{W}=-\frac{j_0}{2}\frac{(G_1\tau/N_0)^2a(a+l^2)}{\left[\left[a+\frac{(b+1)^2}{4b}l^2\right](1+l^2)+d^2\right]^2+\frac{(b-1)^2}{4b}d^2}\ , \tag{48}$$

where the quantities l, a, b, d, and $j_0=j(I_1+I_2)$ are given by Eqs. (33)–(36) and (44). The negative sign for Δj indicates that the photocurrent under an excitation grating, $j(I_1,I_2)$, is always less than or equal to $j(I_1+I_2)$ under a uniform illumination. Intuitively this makes sense because the current flow is limited by regions of lower density of carriers under nonuniform photoexcitation. As expected, Δj vanishes if l and/or d are much greater than unity (smearout of photocarrier gratings due to diffusion and/or drift). It is worth noting that Δj varies linearly with the applied field E_0 only at low E_0 because d and j_0 are both proportional to E_0.

Using Eq. (40) and noting Eqs. (15), (16), and (28)–(31), it can be readily shown that the spatially averaged conductivity $\sigma_{\rm ave}$, in Eq. (39) is

$$\sigma_{\rm ave}=\sigma_0-\frac{e^2}{2\sigma_0}[(\mu_n N_1+\mu_p P_1)^2+(\mu_n N_2+\mu_P P_2)^2]$$
$$=\sigma_0+\frac{\Delta j}{E_0}\left[\frac{a+l^2}{a}\right]\neq\sigma_0+\frac{\Delta j}{E_0}\ , \tag{49}$$

where Δj, given by Eq. (48), is the correct expression for $[j(I_1,I_2)-j(I_1+I_2)]$ derived earlier from the principle of current continuity in the steady state. Equation (49) reveals that $j(I_1,I_2)=\sigma_0E_0+\Delta j\neq\sigma_{\rm ave}E_0$. Clearly, the approach adopted by earlier workers[1–5] to equate $\sigma_{\rm ave}E_0$ and $j(I_1,I_2)$ is appropriate *only if* $a\gg l^2$ (or in practical terms $a\gg 1$ because Δj vanishes when $l\gg 1$), in which case $\sigma_{\rm ave}E_0\approx j_0+\Delta j$ according to Eq. (49). The failure of the "series resistor" model may occur because the SSPG is not a passive system, and carrier diffusion (in addition to drift) can play a role in the macroscopic transport in response to an external field.

In an earlier publication,[5] ambipolar mobility[2,6] was supposedly determined for *a*-Si:H through the analysis of field dependence of $j(I_1,I_2)$ under the erroneous assumption of space-charge neutrality ($\Delta N=\Delta P$). It was argued subsequently that ambipolar mobility would not be vanishing if the small-signal drift mobilities for electrons and holes differ in dissimilar ways from μ_n and μ_p under a uniform excitation.[2] However, the preceding analysis in this manuscript shows that the effect of E_0 is to pull photoelectrons and photoholes in opposite directions so that space charge is produced. Thus the drift motion of electrons and holes, signified by d, is bipolar, not ambipolar (electrons and holes drift together to keep charge neutrality), even if a is large and space-charge density is low (unlike the argument in Ref. 2). Since there is no physical reason for the small-signal mobilities[2] ascribed to ΔN and ΔP to differ greatly from those of the background carriers N_0 and P_0, the derivation of Eq. (48) in this article ought to be principally valid. Therefore, contrary to the reasoning in Ref. 2, the field dependence of Δj (even at low E_0) must be mainly through d or the geometric average of electron and hole mobilities, rather than some speculated ambipolar mobility.[5,2]

To conclude this section, it should be noted that Δj, which can be written as

$$\Delta j\equiv j(I_1,I_2)-j(I_1+I_2)$$
$$=[j(I_1,I_2)-j(I_1)]-[j(I_1+I_2)-j(I_1)]\ , \tag{50}$$

is an experimentally measurable quantity because $[j(I_1+I_2)-j(I_1)]$ and $[j(I_1,I_2)-j(I_1)]$ can be detected, respectively, by chopping the light beam I_2 when I_1 and I_2 do not and do interfere with each other (see Refs.

and 3). Also note that while $j(I_1+I_2)-j(I_1)$ is always positive, $j(I_1,I_2)-j(I_1)$ can be positive or negative[1] depending on l and d. Hence the *phase* of $[j(I_1,I_2)-j(I_1)]$ with respect to that of $[j(I_1+I_2)-j(I_1)]$ needs to be monitored.

IV. DISCUSSION

Information about transport of the less mobile type of photocarriers (holes) is embodied in L and b, which can be probed using Eq. (48). To avoid the difficulties associated with severe space-charge effect, only the situation of $a \geq 1$ will be discussed.

A. The weak-field condition

Previous investigations[1-4] established that L could be deduced from the variation of $j(I_1,I_2)$ (or Δj in this paper) with l by adjusting the grating period Λ. In this kind of experiment, the applied field E_0 should be small to prevent any drift blurring of the photocarrier gratings. An inspection of Eq. (48) suggests that, for any finite b, if the following conditions

$$d^2 \ll a, \quad d^2 \ll l^2 \tag{51}$$

are fulfilled, Eq. (48) can be simplified by dropping the d^2 terms to become

$$\Delta j = -\frac{\Delta j_{\max}\, a(a+l^2)}{\left[\left[a+\frac{(b+1)^2}{4b}l^2\right](1+l^2)\right]^2}, \tag{52}$$

where $\Delta j_{\max}=(G_1\tau/N_0)^2 j_0/2$ is a positive constant (with a fixed E_0) derivable in the limit of $l^2 \ll a$. For semiconductors with low defect density, $a>1$ can be expected as $\tau_{\rm rel}$ can be made small relative to τ under moderate illumination. Note that Δj is most sensitive to the change of l when l is in the neighborhood of unity ($l \sim 1$). Thus, in practical terms, the *weak-field conditions* in Eq. (51) can be approximately stated as $d^2 \ll l^2$ or simply $d \ll l$ for samples with $a>1$ and with a finite b (e.g., $b \leq 20$). According to Eqs. (33) and (36), this condition stipulates

$$E_0 \ll \frac{L}{\tau\sqrt{\mu_n\mu_p}} = \frac{L}{\tau\mu_p\sqrt{b}} = \frac{k_BT}{e}\frac{L}{\tau D_p\sqrt{b}}, \tag{53}$$

where Eq. (20) is used. For $b \gg 1$, Eq. (26) gives $D \approx 2D_p$, and from Eq. (25) one obtains $L^2 \approx 2\tau D_p$. Thus Eq. (53) becomes

$$E_0 \ll \frac{2}{\sqrt{b}L}\frac{k_BT}{e}. \tag{54}$$

For $b>4$, the above requirement is stricter than the one suggested by earlier analyses,[2,4] which specify $E_0 < k_BT/(eL)$ (Refs. 2 and 3) and $E_0 \ll 2\pi k_BT/(e\Lambda)$ (Ref. 4, where $\Lambda \approx 2\pi L$ is considered) to justify the numerical[2] and analytical[4] solutions of the continuity equations obtained by neglecting E_0. Experimentally, the weak-field condition is met if E_0 is well below the onset of sublinearity in the relation of Δj versus E_0 for all the Λ used.

If $a=\tau/\tau_{\rm rel}$ is estimated by measuring τ in the small signal photoconductivity decay experiment,[2,16] Eq. (52) can be employed to deduce L. This is feasible because the factor

$$\frac{(b+1)^2}{4b}l^2 = \frac{(b+1)^2}{4b}(kL)^2 = \frac{\tau(D_n+D_p)}{2}k^2 = \frac{\tau(\mu_n+\mu_p)k_BT}{2e}k^2 \tag{55}$$

in Eq. (52) can be readily computed from the mobility-lifetime[13-17] product, $(\mu\tau)_{\rm pc}$, derived from the steady-state photocurrent $j_{\rm pc}=j_0-j_{\rm dark}$ ($j_{\rm dark}$ is the dark current) according to[17]

$$j_{\rm pc} = eN_{\rm pc}(\mu_n+\mu_p)E_0 = e(\mu\tau)_{\rm pc}G_0E_0, \tag{56}$$

where $N_{\rm pc}$ is the *photocarrier* density. Equation (56) assumes that the illumination is nearly uniform over the volume of the sample, and that the quantum efficiency of photogeneration of electron-hole pairs[13-15] is unity for photons of near-band-gap energies. Since the photocurrent usually shows a power-law dependence on the photon flux or the generation rate G_0, $j_{\rm pc} \propto G_0^{\alpha}$, where α is a constant typically between 0.5 and 1, one has from Eq. (56)

$$N_{\rm pc} \propto G_0^{\alpha}, \tag{57}$$

provided that μ_n and μ_p are insensitive to G_0 (i.e., only τ varies with carrier density). Using Eq. (57) to compute the small-signal τ according to Eq. (12), one finds

$$\tau = \frac{\partial(N_{\rm pc})}{\partial G_0} = \frac{\alpha N_{\rm pc}}{G_0} \tag{58}$$

or $N_{\rm pc}=\tau G_0/\alpha$. Inserting this into Eq. (56), one gets

$$\tau(\mu_n+\mu_p) = \alpha(\mu\tau)_{\rm pc} = \frac{\alpha j_{\rm pc}}{eG_0E_0}, \tag{59}$$

i.e., $(\mu\tau)_{\rm pc}$ as conventionally defined[17] in Eq. (56) differs by a factor of α from the product of $(\mu_n+\mu_p)$ and τ used in Eqs. (13) and (14), because τ is not the same quantity in $(\mu\tau)_{\rm pc}$ unless $\alpha=1$. One concludes from the procedure outlined above that L can be determined even if the lifetime regime[2] or ambipolarity[4] requirement ($a>b$) is *not* met. Note that $\Delta j_{\max}$ in Eq. (52) can be eliminated in the analysis of Δj versus l by taking the *ratios* of Δj measured at different Λ under a fixed E_0. In this way, L can be sought even if the dark current is high.[10] This is different from the standard routine[1-4] (best suited for photoconductive insulators) of taking the ratio of $[j(I_1,I_2)-j(I_1)]$ to $[j(I_1+I_2)-j(I_1)]$. In passing, it is worth remarking that, if τ (and hence a) is unknown, one may still be able to use Eq. (52) to explore both a and L. In this scheme, one would search for a best fit to the data of Δj versus Λ by adjusting the two parameters a and L ($l=2\pi L/\Lambda$) in Eq. (52). The uniqueness of such a fit needs to be tested.

In the so-called lifetime regime,[2] $\tau \gg \tau_{\rm rel}$ (e.g., for device quality a-Si:H[2]), such that $a \gg bl^2/4$. For l near unity, if $a>b$ or

$$\frac{\tau}{\tau_{\rm rel}} > \frac{\mu_n}{\mu_p} \tag{60}$$

holds, space-charge neutrality is well kept ($\Delta N \approx \Delta P$, the gratings of photoelectrons and photoholes essentially overlap) and ΔE is negligible, as can be demonstrated by examining Eqs. (28)–(31), (37), and (38). In the lifetime regime, Eq. (52) can be reduced to show a simple dependence of Δj on l,

$$\Delta j \approx -\Delta j_{\rm max}(1+l^2)^{-2} = -\frac{j_0}{2}\left[\frac{G_1\tau}{N_0}\right]^2(1+l^2)^{-2} . \tag{61}$$

For a *photoconductive insulator* whose dark carrier density is low, one has $N_0 \approx N_{\rm pc} = \tau G_0/\alpha$ by virtue of Eq. (58). Inserting this into Eq. (61) one finds

$$\Delta j \approx -\frac{j_0}{2}\left[\alpha\frac{G_1}{G_0}\right]^2(1+l^2)^{-2} , \tag{62}$$

which can easily be shown to be identical to the formula derived earlier[1,3] under the assumption of space-charge neutrality in a weak field. Formula (62) has been much used in extracting L from a-Si:H and its alloys.[18] The validity condition ($a > b$) for Eq. (61) or (62) was previously indicated by numerical solutions of the continuity equations.[2] It has been established by numerical simulations[2] and by comparing L derived from Eqs. (52) and (62) using wide-ranging values for a and b that, if $a < b$, an overestimate of L will result[10] by adopting Eq. (62).

B. The strong-field condition

There exists the possibility that b can be inferred from the field dependence of Δj when the diffusion terms in Eq. (48) are negligible, i.e., when $a \gg bl^2/4$ and $d^2 \gg l^2$. The first condition is easily satisfied for the case of $a > 1$ by choosing large Λ so that $l^2 \ll 1$. The second *strong-field condition* is just the case opposite to the previously discussed weak-field condition ($d^2 \ll l^2$) expressed in Eq. (54). Hence when

$$E_0 \gg \frac{2}{\sqrt{b}L}\frac{k_BT}{e} \tag{63}$$

holds, Eq. (48) can be written as (recall $j_0 \propto E_0$)

$$\frac{\Delta j}{E_0} = -\frac{\Delta\sigma_{\rm max}a^2}{(a+d^2)^2+\dfrac{(b-1)^2}{4b}d^2} , \tag{64}$$

where $\Delta\sigma_{\rm max} = \Delta j_{\rm max}/E_0$ is a constant that can be established at sufficiently low E_0 such that $d^2 \ll a$. Because of the following relation

$$d^2 = k^2(\tau\sqrt{\mu_n\mu_p}E_0)^2 = \frac{k^2b}{(b+1)^2}[\tau(\mu_n+\mu_p)E_0]^2 , \tag{65}$$

where $(\mu_n+\mu_p)\tau$ can be calculated using Eq. (59) from the background photocurrent, Eq. (64) contains only two independent unknowns, a and b. Should a be determined through the measurements of τ, b can be deduced by analyzing the variation of $\Delta j/E_0$ with E_0 under strong E_0 (e.g., d^2 near a) with a fixed grating period Λ. Once b is obtained, the hole diffusion length can be estimated from the electron diffusion length (the two differ by a factor of $b^{1/2}$) attainable from steady-state photoresponse $(\mu\tau)_{\rm pc}$. It appears that the problem of deducing b becomes much more challenging when a is not known (the photocurrent initial-decay experiment[2,16] for determining τ is by no means trivial). It is of interest to find out whether or not *both* a and b can be found simultaneously by using a and b as adjustable parameters in Eq. (64) to search for the best fit to the data of $\Delta j/E_0$ versus E_0. An answer of yes to this question would prove useful for semiconductors with small L that may not be measurable using the weak-field procedure outlined earlier, as there exists an empirical detection limit for L to about a few hundred Å using the SSPG technique.[2,19]

V. SUMMARY

This paper has shown that, under the presence of a small optical excitation grating in a *finite* electric field, the continuity equations can be linearized and solved *analytically* provided that the electric field grating is small compared with the thermal voltage divided by the illumination grating period Λ. The general formulas for photocarrier densities and transport can be expressed in terms of several dimensionless, physically meaningful ratios: the photocarrier recombination lifetime τ to dielectric relaxation time $\tau_{\rm rel}$($a = \tau/\tau_{\rm rel}$), the electron mobility to hole mobility ($b = \mu_n/\mu_p$), the ambipolar diffusion length L to Λ ($l = 2\pi L/\Lambda$), and the mean drift length of electrons and holes $(\mu_n\mu_p)^{1/2}\tau E_0$ to Λ [$d = 2\pi(\mu_n\mu_p)^{1/2}\tau E_0/\Lambda$]. Charge continuity analysis has revealed that the series-resistor model in computing photoconductivity[1–4] is appropriate only if $a \gg l^2$. For the SSPG technique to be usable for determining L or b, a should be near to or greater than unity so that space-charge effect is not too severe to satisfy the linearization condition for a finite excitation grating. In the weak-field case, the lifetime regime[2] or ambipolarity[4] ($a > b$) requirement is shown to be favorable but *unnecessary* for the determination of L, especially when τ is measured. Only when a is quite high can the SSPG problem be treated by assuming space-charge neutrality,[1] in which case the general transport formula derived here is reduced to the more familiar, much used simple formula.[1] The possibility exists that, by studying the dependence of photocurrent on the applied field, one may be able to deduce b and get an idea about hole transport.

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*Present address: Thin Film Division, Solarex Corporation, 826 Newtown-Yardley Road, Newtown, PA 18940.

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