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Ambipolar transport in amorphous semiconductors in the lifetime and relaxation-time regimes investigated by the steady-state photocarrier grating technique

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The ambipolar-transport equations including space-charge effects are solved for the case of a sinusoidal generation of photocarriers in amorphous semiconductors. In the "lifetime regime" where the dielectric relaxation time is much shorter than the lifetime no space-charge effects exist, i.e., electrons and holes move together even if their mobilities are different and an electric field is applied. In the "relaxation regime," where the opposite relation between lifetime and relaxation time prevails, separation of electrons and holes occurs for different mobilities of the carriers. In any case, an electric field will separate the carriers in this regime. We apply the theory to examine experimental results for the diffusion length of photocarriers in a sample of hydrogenated amorphous silicon obtained by the steady-state photocarrier grating technique. We find that space-charge effects are not serious at low electric fields so that the true ambipolar diffusion length is obtained, but that separation of charges occurs at high electric fields.

I. INTRODUCTION

Recently, the authors have introduced a steady-state photocarrier grating technique (SSPG) for diffusion-length measurement in semiconductors[1–4] and have applied it to hydrogenated amorphous silicon (a-Si:H) and semi-insulating gallium arsenide. In the original papers on the subject, as well as in other papers dealing with diffusion-length measurements by other methods,[5] only the "lifetime regime"[6–12] for the excess carriers was considered. This regime refers to the case where the lifetime of excess carriers is much longer than the dielectric relaxation time of the material so that charge neutrality prevails everywhere and electrons and holes diffuse together. In this paper we also consider the "relaxation-time regime"[6–12] in which the opposite relation between the lifetime and dielectric relaxation time prevails. In this case local charge neutrality is not preserved and electrons and holes can be spatially separated. For a finite ratio of lifetime to dielectric relaxation time separation of charges will always set in at high enough electric fields.

In Sec. II we first present a phenomenological approach which enables us to treat ambipolar transport in a very general manner. Then, in Sec. III, we consider the case of ambipolar transport when charge neutrality is maintained. This regime is of interest since, as demonstrated in this paper, space-charge effects can usually be neglected at low external electric fields, in the case of a-Si:H. In Sec. IV space-charge effects are incorporated into the theory of the SSPG experiment, and we distinguish between two cases: space-charge effects due to unequal diffusion constants of electrons and holes, and space-charge effects caused by an external electric field. It is found that the transition to the relaxation regime mainly depends on the ratio between the dielectric relaxation time and the carrier lifetime, and the ratio between the electron and hole drift mobilities. Space-charge effects caused by an external electric field are found to be more pronounced than space-charge effects due to the different values of the diffusion constants of electrons and holes. In Sec. V experimental results for the small-signal lifetime of photocarriers in a-Si:H are presented. It is demonstrated that in the SSPG diffusion-length measurement at low electric fields electrons and holes diffuse together, i.e., charge neutrality is maintained, but that when an electric field is applied electrons and holes are separated and charge neutrality is not maintained. Previous results of the electric field dependence of the photocarrier grating amplitude[4] are shown to be due to space-charge effects.

II. A PHENOMENOLOGICAL APPROACH TO AMBIPOLAR TRANSPORT IN AMORPHOUS SEMICONDUCTORS

A. Definition of the transport parameters

Since the microscopic transport and recombination mechanisms in amorphous semiconductors are yet not well understood, we describe the ambipolar transport of photocarriers in a phenomenological manner which does not depend on the details of the transport and recombination mechanisms. In this approach the concept of drift mobility is used which is an average mobility of all carriers involved in the transport and is thus model independent. A further advantage of such an approach is that space-charge effects, which are due to both mobile and trapped carriers, are taken into account in a natural manner.

The drift mobility of electrons, μ_n, and that of holes, μ_p, is defined by the relations[13]

$$\mathbf{J}_n^{\mathrm{drift}} = -\mathbf{E}\mu_n N \ , \tag{1a}$$

$$\mathbf{J}_p^{\mathrm{drift}} = \mathbf{E}\mu_p P \ , \tag{1b}$$

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where N and P are the total, mobile and trapped, electron and hole concentrations, $\mathbf{J}_n$ and $\mathbf{J}_p$ the electron and hole fluxes, and $\mathbf{E}$ the electric field. In a self-consistent manner, the diffusion currents are described with use of effective diffusion constants of electrons and holes:

$$\mathbf{J}_n^{\text{diff}} = -D_n \nabla N \ , \tag{2a}$$

and

$$\mathbf{J}_p^{\text{diff}} = -D_p \nabla P \ . \tag{2b}$$

We now evaluate the generalized Einstein relationship between the electron drift mobility and effective diffusion constant:[14]

$$-\mu_n N = qD_n \frac{dN}{d\varepsilon} = qD_n \int_{-\infty}^{\infty} g(\varepsilon) \frac{df(\varepsilon)}{d\varepsilon} d\varepsilon \ . \tag{3}$$

Here $g(\varepsilon)$ is the density of states of electrons, $f(\varepsilon)$ is the occupancy function, N the total concentration of electrons, and q the electron charge.

The electron occupancy function for photo-excited carriers was shown by Rose[15] to be determined by thermal equilibration for states which are located above the quasi-Fermi-level (trap states), and by a dynamic equilibrium for states located well below the quasi-Fermi-level (recombination centers). We thus conclude that $f(\varepsilon)$ is the Fermi-Dirac distribution function for energies above the Fermi level, ε_{fn}, but may differ from the Fermi-Dirac distribution function below ε_{fn}. The derivative of $f(\varepsilon)$ well below ε_{fn} should be zero just as for the Fermi-Dirac function since $f(\varepsilon)$ should be constant within a band of recombination centers. The derivative will also go to zero several kT's above ε_{fn} where $f(\varepsilon)$ becomes a Boltzmann distribution. The main contributions to the integral in Eq. (3) come from energies near ε_{fn} as long as $g(\varepsilon)$ does not increase more rapidly than $f(\varepsilon)$ decreases above ε_{fn}. To evaluate the integral we therefore approximate $g(\varepsilon)$ next to ε_{fn} by the first term in the Taylor-series expansion of the logarithmic density of states:

$$g(\varepsilon) = g(\varepsilon_{fn}) \exp[\alpha(\varepsilon - \varepsilon_{fn})/kT] \ , \tag{4}$$

where k is Boltzmann's constant and T the temperature. The bandtail parameter α is to be thought of as characterizing $g(\varepsilon)$ near ε_{fn} and may well be different at other energies. The only restriction on it is that it should be in the range from zero to unity but not close to either value so that $g(\varepsilon)f(\varepsilon)$ vanishes at $\varepsilon_{fn} \pm \Delta\varepsilon$, where $\Delta\varepsilon$ is several kT's. With this restriction Eq. (3) can be integrated by parts[16] to yield

$$-u_n N = qD_n \int_{\varepsilon_n - \Delta\varepsilon}^{\varepsilon_{fn} + \Delta\varepsilon} \frac{dg(\varepsilon)}{d\varepsilon} f(\varepsilon) d\varepsilon = -qD_n \alpha N/kT \tag{5}$$

with the result

$$D_n = \frac{kT}{\alpha q} \mu_n \ , \tag{6a}$$

and similarly for holes

$$D_p = \frac{kT}{\beta q} \mu_p \ , \tag{6b}$$

where β/kT is the exponent which describes the hole density of states next to the quasi-Fermi-level of the holes.

B. The small-signal approach

Both the drift mobility and the effective diffusion constant are average parameters of all carriers, and may thus depend on the carrier concentration. To avoid a probable nonlinear nature of the transport equations due to the dependence of the transport parameters on the carrier concentrations, we adopt here the small-signal approach as used in Refs. 1–5. In this approach a small nonuniform carrier concentration is superimposed on top of a large uniform carrier concentration that sets the values of the transport parameters. As a result, linear transport equations are obtained.

We define the small-signal electron drift mobility μ_n' by the equation

$$\Delta\mathbf{J}_n = -\mathbf{E}\mu_n' \Delta N \ , \tag{7}$$

where $\Delta\mathbf{J}_n$ is a small change in the electron flux caused by a small perturbation ΔN of the total electron concentration. From Eq. (1) μ_n' is given by

$$\mu_n' = \mu_n + \frac{d\mu_n}{dN} N \ , \tag{8a}$$

and similarly the hole small-signal drift mobility is

$$\mu_p' = \mu_p + \frac{d\mu_p}{dP} P \ . \tag{8b}$$

In order to obtain the relationship between μ and μ' we assume that only a fraction of the total carrier concentration N is mobile and the rest of the carriers are trapped. We denote the mobile carrier concentration by N_m, and assign to these carriers a microscopic mobility μ^0. These assumptions clearly hold within the free-carrier transport model,[17] and have also been shown to be a good approximation for bandtail hopping transport.[18,19] Within this model one writes the drift mobility as[13]

$$\mu_n = \frac{N_m}{N} \mu^0 \ , \tag{9}$$

and it is readily verified that

$$\mu_n' = \mu^0 \left[\frac{N_m}{N} + N \frac{d}{dN} \left[\frac{N_m}{N} \right] \right] = \frac{dN_m}{dN} \mu^0 \ . \tag{10}$$

Equation (10) can now be written in the form

$$\mu_n' = \mu^0 \frac{dN_m/d\varepsilon}{dN/d\varepsilon} = \mu^0 \frac{-(1/kT)N_m}{-(\alpha/kT)N} = \mu_n/\alpha \ . \tag{11a}$$

The above result was obtained by using Eqs. (3) and (5) for $dN/d\varepsilon$ and by assuming that the mobile carriers obey Boltzmann statistics. Equivalently, for holes one has

$$\mu_p' = \mu_p/\beta \ . \tag{11b}$$

Combining Eqs. (6) and (11) we thus obtain the conventional Einstein relationship between the small-signal drift mobility and the effective diffusion constant:

$$D_n=\frac{kT}{q}\mu_n' \quad (12a)$$

and

$$D_p=\frac{kT}{q}\mu_p' . \quad (12b)$$

Note that the above relationship holds in the general case regardless of the value of the integral in Eq. (5), as is evident from Eqs. (3) and (11).

In our small-signal approach, the recombination rate is also formulated in a phenomenological manner to avoid being restricted to a specific recombination mechanism. Most generally, the recombination rate is given by some function of the electron and hole concentrations, $R(N,P)$, and the change in the recombination rate, ΔR, is thus given by

$$\Delta R=\frac{\partial R}{\partial N}\Delta N+\frac{\partial R}{\partial P}\Delta P\equiv\frac{\Delta N}{2\tau_n}+\frac{\Delta P}{2\tau_p} . \quad (13)$$

The electron and hole recombination times, τ_n and τ_p, as defined by Eq. (13) are the average lifetimes of free and trapped carriers and should not be confused with the free-carrier lifetimes. When charge neutrality is maintained $\Delta N=\Delta P$, and a common recombination time may be defined:

$$\Delta R=\Delta N(1/2\tau_n+1/2\tau_p)\equiv\Delta N/\tau . \quad (14)$$

The common recombination time τ can be shown to be equivalent to the photocurrent response time if release times from traps are shorter than τ, as discussed in the Appendix.

III. AMBIPOLAR TRANSPORT IN THE LIFETIME REGIME

A. The ambipolar-transport equation

As shown below, local charge neutrality is frequently maintained at low electric fields and we therefore first discuss ambipolar transport in the lifetime regime. We shall derive an expression for the ambipolar diffusion constant which is more general than that derived by Moore,[5] and also derive an expression for the ambipolar mobility which has not been previously given.

The continuity equations for the electron and hole currents are given by[20]

$$-\nabla\cdot(-\mathbf{E}\mu_n N-D_n\nabla N)+G-R=0 \quad (15a)$$

and

$$-\nabla\cdot(\mathbf{E}\mu_p P-D_p\nabla P)+G-R=0 , \quad (15b)$$

where both the drift mobility and diffusion constants are functions of the carrier concentrations. For a small perturbation ΔG in the otherwise uniform generation rate one has

$$\mu_n N\nabla\cdot\mathbf{E}+\mu_n'\mathbf{E}\cdot\nabla(\Delta N)+D_n\nabla^2(\Delta N)+\Delta G-\Delta R=0 , \quad (16a)$$

and

$$-\mu_p P\nabla\cdot\mathbf{E}-\mu_p'\mathbf{E}\cdot\nabla(\Delta P)+D_p\nabla^2(\Delta P)+\Delta G-\Delta R=0 , \quad (16b)$$

where second-order terms in ΔN and ΔP have been omitted. In the lifetime regime $\Delta N=\Delta P$ locally, and the two equations can be combined in the usual manner[20] into the single ambipolar-transport equation:

$$\mu\mathbf{E}\cdot\nabla(\Delta N)+D\nabla^2(\Delta N)-\Delta N/\tau+\Delta G=0 . \quad (17)$$

The ambipolar diffusion constant D is found to be

$$D=\frac{D_n\mu_p+D_p\mu_n}{\mu_n+\mu_p}=(kT/q)\mu_p\frac{b(1/\alpha+1/\beta)}{b+1} , \quad (18)$$

where $b=\mu_n/\mu_p$ and use has been made of Eq. (6). Equation (18) differs from that obtained by Moore[5] because of the terms involving α and β. Since, as discussed above, we expect these parameters to be between zero and unity but not close to either value, the ambipolar diffusion constant will be determined by the carrier with the lower drift mobility.

The ambipolar drift mobility μ is found to be

$$\mu=\frac{\mu_n'\mu_p-\mu_p'\mu_n}{\mu_n+\mu_p}=\mu_p\frac{b(1/\alpha-1/\beta)}{b+1} . \quad (19)$$

Note that the ambipolar mobility is not zero as would be the case for an intrinsic crystalline semiconductor unless, fortuitously, $\alpha=\beta$. The "Einstein relationship" between the ambipolar mobility and ambipolar diffusion constant thus becomes

$$\frac{D}{\mu}=\frac{kT}{q}\frac{\alpha/\beta+1}{\alpha/\beta-1} . \quad (20)$$

It must be borne in mind, however, that the concept of ambipolar mobility is only valid in the lifetime regime, when charge neutrality is maintained. This is not the case when strong electric field is applied, as discussed below.

B. The relationship between the ambipolar diffusion length and the photoconductivity

In this section we establish the relationship between the ambipolar diffusion length $L=\sqrt{D\tau}$ and the photoconductivity in the same sample. Within the small-signal approach we first find the expression for a change in the conductivity due to a small change, ΔG, in the generation rate. From Eq. (7) one obtains

$$\Delta\sigma=q(\mu_n'\Delta N+\mu_p'\Delta P) , \quad (21)$$

and since in the steady state $\Delta R=\Delta G$, one has from Eq. (14)

$$\Delta\sigma=q\,\Delta G\,\tau(\mu_n'+\mu_p')=q\,\Delta G\,\tau(\mu_n/\alpha+\mu_p/\beta) . \quad (22)$$

From Eqs. (18) and (22) one obtains

$$\frac{L^2}{\Delta\sigma/(q\,\Delta G)}=\frac{kT}{q}\frac{b}{b+1}\frac{\alpha/\beta+1}{\alpha/\beta+b} . \quad (23)$$

Equation (23) may thus be employed in order to obtain an approximate value of b from a measurement of the

small-signal photoconductivity and the diffusion length in the same sample if it is assumed that α/β is of order unity and no space-charge effects exist at low electric fields. We show in Sec. VI that for a representative sample of a-Si:H both of these assumptions are justified.

IV. AMBIPOLAR TRANSPORT IN THE RELAXATION REGIME

A. Solution of the SSPG transport equations

In the dark, typical amorphous semiconductors such as a-Si:H are clearly "relaxation semiconductors"[6] since their dielectric relaxation time $\tau_d=\kappa\kappa_0/\sigma$ is very long because of their low conductivity σ. When illuminated, however, the conductivity may rise by several orders of magnitude[13] and τ_d will correspondingly decrease. It is therefore not clear, *a priori,* whether the lifetime regime or the relaxation-time regime applies to the ambipolar transport of excess carriers under illumination. In this section the solutions of the transport equations relevant to the SSPG experiment are presented, and the transition from the lifetime regime to the relaxation regime is investigated.

In the SSPG experiment,[1–4] the generation rate G is composed of a small sinusoidal part superimposed on a uniform generation rate:

$$G=G_0+G_1\cos(kx) , \tag{24}$$

where $G_1 \ll G_0$, $k=2\pi/\Lambda$, and Λ is the grating period. The electron concentration N and the hole concentration P are also sinusoidal in space and are given by

$$N=N_0+\mathrm{Re}[N_1\exp(ikx)] , \tag{25a}$$

$$P=P_0+\mathrm{Re}[P_1\exp(ikx)] , \tag{25b}$$

where $N_0=P_0$ are the electron and hole concentrations generated by G_0, and N_1 and P_1 are complex numbers that give the amplitudes and phases of the sinusoidal part of the concentrations. The solution of the SSPG transport equations is greatly simplified by the sinusoidal form of the carrier concentrations, given in Eq. (25). Instead of solving three coupled differential equations one obtains a simple set of three linear equations. The electric field is written as

$$E=E_0+\mathrm{Re}[E_1\exp(ikx)] , \tag{26}$$

and the Poisson equation thus becomes

$$i\kappa\kappa_0 kE_1=q(P_1-N_1) , \tag{27}$$

where κ is the permittivity of the material. The small-signal recombination rate is also sinusoidal:

$$\Delta R=\mathrm{Re}[R_1\exp(ikx)] , \tag{28}$$

and the linear continuity equations that are solved together with the Poisson equation (27) thus become

$$i\mu_n N_0 kE_1+i\mu_n' E_0 kN_1-D_n k^2 N_1+G_1-R_1=0 \tag{29a}$$

and

$$-i\mu_p P_0 kE_1-i\mu_p' E_0 kP_1-D_p k^2 P_1+G_1-R_1=0 . \tag{29b}$$

From Eqs. (13), (25), and (28) we write the recombination term as

$$R_1=N_1/2\tau_n+P_1/2\tau_p , \tag{30}$$

where τ_n and τ_p are coupled according to Eq. (14):

$$1/2\tau_n+1/2\tau_p=1/\tau , \tag{31}$$

and τ is the recombination time discussed above.

We finally introduce the dielectric relaxation time τ_d:

$$\tau_d=\kappa\kappa_0/[q(\mu_n N_0+\mu_p P_0)] . \tag{32}$$

Note that the photoconductivity determines τ_d. Using Eqs. (24)–(32) two coupled linear equations for the unknown concentration N_1 and P_1 are obtained:

$$\begin{aligned} G_1\tau&=P_1[\eta-cb/(1+b)] \\ &\quad +N_1[cb/(1+b)-ikbrL_{ep}+k^2brL_{dp}^2+\delta] , \\ G_1\tau&=P_1[c/(b+1)+ikL_{ep}+k^2L_{dp}^2+\eta] \\ &\quad +N_1[\delta-c/(1+b)] , \end{aligned} \tag{33}$$

where $L_{ep}=\mu_p' E\tau$ is the hole drift length, $L_{dp}=(D_p\tau)^{1/2}$ is the hole diffusion length, and $N_0=P_0$. The dimensionless constants in these equations are

$$b=\mu_n/\mu_p,\quad c=\tau/\tau_d,\quad r=\beta/\alpha$$

and

$$\eta=\tau/2\tau_p,\quad \delta=\tau/2\tau_n,\quad \text{where } \eta+\delta=1 .$$

Equation (33) is very general, and is not restricted to any specific transport model.

We now examine solutions of Eq. (33) for several cases. In order to obtain physical insight into the behavior of the excess carrier we shall, for the moment, assume that $\alpha=\beta$ and that $\tau_n=\tau_p$. The latter assumption has only a minor effect on the solutions since τ_n and τ_p are coupled via Eq. (31). The assumption that $\alpha=\beta$ has a more serious effect for the case of a nonzero electric field, as is evident from the expression for the ambipolar mobility given in Eq. (19). The case when $\alpha=\beta$ is of interest, however, since the ambipolar mobility then equals zero, and the effect of the electric field is solely due to the finite value of τ/τ_d, namely the transition to the dielectric relaxation regime.

B. Zero electric field

When no external electric fields are applied, space charges can only be created due to faster diffusion of one type of carriers. The solution of Eq. (33) for $E=0$ and $\Lambda=10L$ is shown in graphical form in Fig. 1 where the normalized amplitudes of the electron and hole gratings are plotted versus the ratio τ/τ_d. Note that when τ/τ_d is large electrons and holes diffuse together to maintain charge neutrality even if $b\neq 1$. In this case, $N_1=P_1$ and one obtains

$$N_1=P_1=G_1\tau/[1+(2\pi L/\Lambda)^2] , \tag{34}$$

where L is the ambipolar diffusion length. This relation

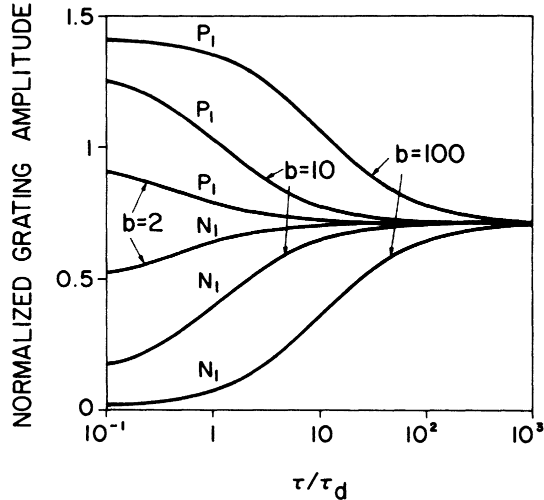

FIG. 1. Theoretical plot of the electron and hole grating amplitudes N_1 and P_1 at $E=0$, vs τ/τ_d. The amplitudes are normalized by $G_1\tau$ which is their value in the absence of diffusion. The parameter b is the ratio μ_n/μ_p, and the grating period Λ was chosen to equal $10L$.

has been used by the authors previously in order to describe SSPG experiments at low electric fields[1-4] and is thus justified in the lifetime regime.

When τ/τ_d is small, local charge neutrality is no longer maintained and electrons and holes diffuse independently if $b\neq 1$. In this regime the electron grating amplitude decreases due to faster diffusion of the electrons. The resulting decrease of the recombination rate leads to an increase in the hole grating amplitude. The transition between the lifetime regime, where charge neutrality is maintained, and the relaxation regime, where electrons and holes can be separated by diffusion, is seen to depend on the ratio $b=\mu_n/\mu_p$ and to occur roughly when $\tau/\tau_d=b$.

The practical implications for SSPG measurements even at negligible electric fields are immediately obvious. In the SSPG method[1-4] a sinusoidal conductivity pattern of the form

$$\sigma=\sigma_0+\mathrm{Re}[\sigma_1\exp(ikx)] \qquad (35)$$

is generated in the sample. In Eq. (35) $\sigma_0=q(\mu_n+\mu_p)N_0$ is the conductivity generated by the uniform background illumination, and the complex amplitude of the conductivity grating is given by

$$\sigma_1=q(\mu_n'N_1+\mu_p'P_1)\ . \qquad (36)$$

Comparing Eq. (36) to Eq. (22), a normalized conductivity grating amplitude γ can be defined by the expression

$$\gamma=\frac{|\sigma_1|}{q(\mu_n'+\mu_p')G_1\tau}\ . \qquad (37)$$

The normalized conductivity grating amplitude equals unity in the case of negligible diffusion and drift, and is reduced if blurring of the grating by diffusion or drift sets in. As described in Ref. 2, in order to obtain the ambipolar diffusion length L, γ is measured at low electric fields for several grating periods, and Eq. (34) is then used to calculate L. Since Eq. (34) is valid only if the charge neutrality is maintained, the result will clearly be different for the case of a single sinusoidal variation for both electrons and holes and the case of two different sinusoidal curves for the two types of carriers. To find out under what conditions the use of Eq. (34) is justified, namely when the transition to the relaxation regime occurs, we have calculated γ as a function of $c=\tau/\tau_d$ for various values of $b=\mu_n/\mu_p$. The ratio between the apparent diffusion length, L_{app}, which is obtained using Eq. (34), and the "true" ambipolar diffusion length is found for various values of b and c. The results are given in Fig. 2 where we plot the ratio $L_{\mathrm{app}}^2/D\tau$ for different values of b and c. Clearly, this ratio equals unity if $b=1$ since electrons and holes then have the same diffusion constants and charge neutrality is always maintained, or if τ/τ_d is sufficiently large to prevent space charge even if $b\neq 1$.

Since one does not know the values of b and c to start with it may at first glance be impossible to draw conclusions about L from a SSPG measurement even at low electric fields. As shown below, however, we have found that in a representative sample of a-Si:H lifetime regime conditions prevail at $E=0$.

C. The case of an external electric field

We now turn to a solution, Eq. (33), for the case of $E>0$ and display the results in graphic form in Fig. 3. In solving these equations we have assumed that $\alpha=\beta$ so that the ambipolar drift mobility, as given by Eq. (19), is zero. When an external electric field is applied the transition from the lifetime regime to the relaxation regime occurs at lower values of the ratio τ/τ_d than in the case of $E=0$. This can be seen in Fig. 3 where the electron

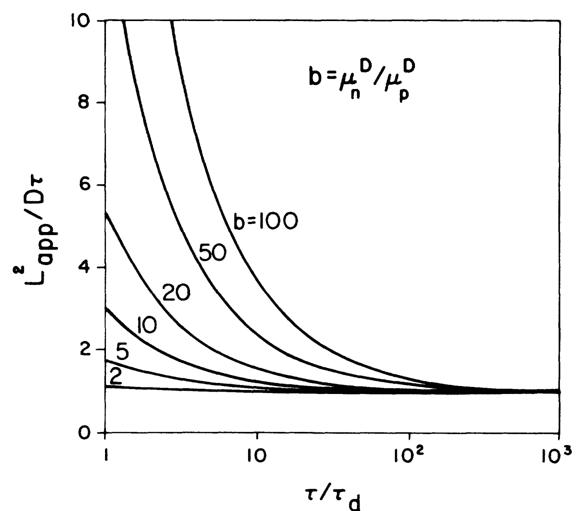

FIG. 2. Theoretical plot of the ratio $L_{\mathrm{app}}^2/D\tau$ vs the ratio between the lifetime τ and the dielectric relaxation time τ_d. L_{app} is the apparent ambipolar diffusion length obtained from the SSPG diffusion-length measurement if space-charge effects are not taken into account. D is the ambipolar diffusion constant and τ the carrier lifetime.

and hole grating amplitudes are plotted versus τ/τ_d with an electric field of magnitude $\Lambda/2\pi\mu_p\tau$ applied in the x direction. In the lifetime regime the application of an electric field does not reduce the amplitude of the carrier grating as is expected since we have assumed zero ambipolar drift mobility. In the relaxation regime, however, the electric field separates electrons and holes, as can be seen from the opposite phase shifts of the two gratings. In this case the grating amplitudes are determined by independent drift of electrons and holes, and subsequent changes in the recombination rate. The transition to the relaxation regime can be seen to occur roughly when $\tau/\tau_d = 10b$.

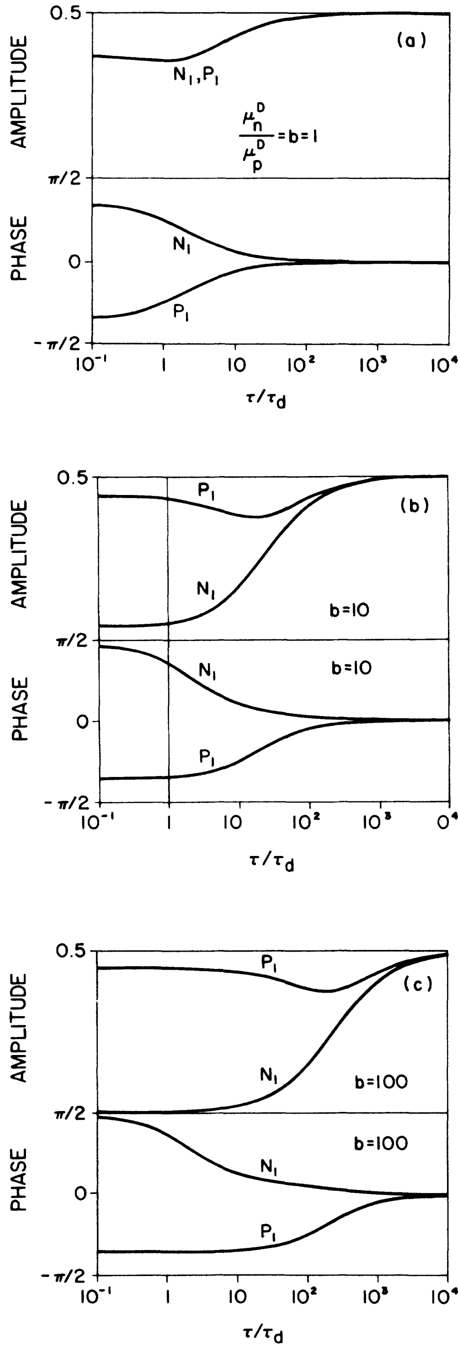

FIG. 3. Theoretical plot of the electron and hole grating amplitudes and phases vs τ/τ_d for $E = \Lambda/\pi\mu_p\tau$. The amplitudes are normalized by $G_1\tau$ which is their value in the absence of the drift and diffusion. The grating period Λ equals $2\pi L$, where L is the ambipolar diffusion length. The parameter b is the ratio between the electron and hole drift mobilities, which is taken as (a) 1, (b) 10, and (c) 100.

In order to compare these theoretical results to the experimental data in the next section we calculated the normalized conductivity grating amplitude γ versus the applied external field. We again let $\alpha = \beta$ so that the ambipolar mobility vanishes. The conductivity grating amplitude is then calculated from Eq. (36) by solving Eq. (33) for N_1 and P_1 as a function of the external electric field. The results are presented in Fig. 4. For the sake of illustration we have assumed equal electron and hole drift mobilities. As expected, when τ/τ_d is large there is practically no effect of the applied electric field on the conductivity grating amplitude. For smaller values of τ/τ_d, on the other hand, the field blurs the grating. The blurring of the conductivity grating is due to the reduction of the electron and hole grating amplitudes described above, and also to the phase shift between them, since the conductivity is an averaged sum of both concentrations.

V. COMPARISON WITH EXPERIMENTAL RESULTS

A. Determination of the carrier lifetime in a-Si:H

The ratio between the carrier lifetime τ and the dielectric relaxation time τ_d must be known in order to apply the theory to experimental data. We have measured the carrier lifetime in a representative sample of a-Si:H and then used the result in order to obtain exact solutions to the continuity equations. A 3-μm-thick sample of a-Si:H deposited by glow discharge decomposition of silane on a quartz substrate was used in these experiments. The contacts were coplanar chromium electrodes with a gap of

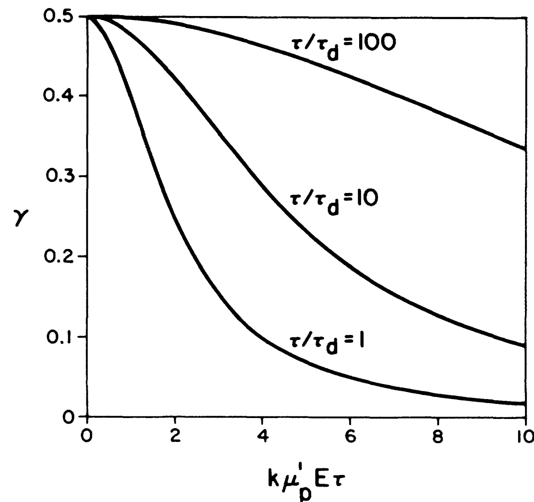

FIG. 4. Theoretical graph of the normalized conductivity grating amplitude as defined in Eq. (37), vs $k\mu_p'E\tau$, for $\mu_p = \mu_n$, and $\alpha = \beta$. Here $k = 2\pi/\Lambda$, and the grating period Λ is chosen to equal $2\pi L$.

0.3 mm deposited on top of the sample.

We measured the carrier lifetime τ from the small-signal photocurrent decay time. As is further discussed in the Appendix, when the photocurrent decays exponentially, the decay time constant is the carrier lifetime defined in Sec. II. The sample was illuminated by a He-Ne laser with an average intensity of 70 mW/cm^2. This background illumination is the same as in the photoconductivity and diffusion-length measurements which were also carried out on the same sample. About 5% of the intensity of the beam was modulated by a square waveform at about 100 Hz using an electrooptic modulator, and the photocurrent decay was monitored by a transient digitizer. Care was taken to insure that the RC response time of the circuit is smaller than the photocurrent response time. The results are given in Fig. 5. The small-signal photocurrent transient does indeed decay exponentially, and the time constant of the decay, which is also the carrier lifetime, equals 1.8 μsec.

Since the decay rate strongly depends on light intensity, and since definitions of the lifetime vary in various publications, we do not attempt to compare the value of τ found by us to values of the lifetime in the literature.[21-24] It is shown in the Appendix, though, that the measured τ is the lifetime defined in the theoretical selections of this paper, provided tha the bias light intensity is the same in all experiments.

B. The effect of an external field on the conductivity grating amplitude

In this section we reinterpret the results of the SSPG experiment carried out as a function of an external electric field.[4] Details of the experiment were given in Ref. 4 but in contrast to the interpretation of the results in Ref. 4 we now do not assume local charge neutrality but take space-charge effects into account.

The experimental results are given in Fig. 6. The normalized conductivity grating amplitude γ was measured at a bias light intensity of 70 mW/cm^2 using the method described in detail in Ref. 2. The amplitude of the grating at low electric fields is determined by diffusion only, whereas, as shown above, at higher electric fields the grating amplitude decrease due to an additional drift of carriers.

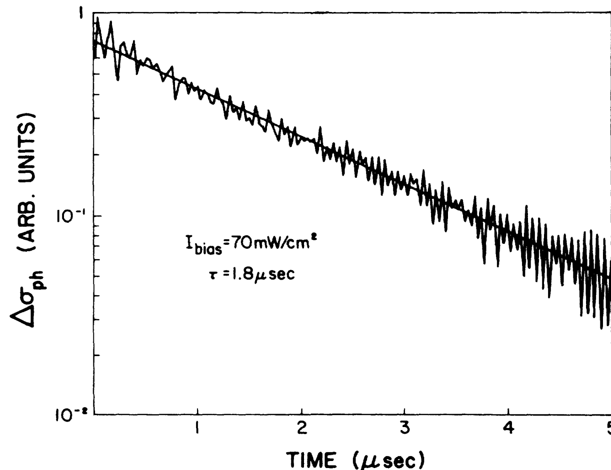

FIG. 5. Small-signal photocurrent decay from the steady state.

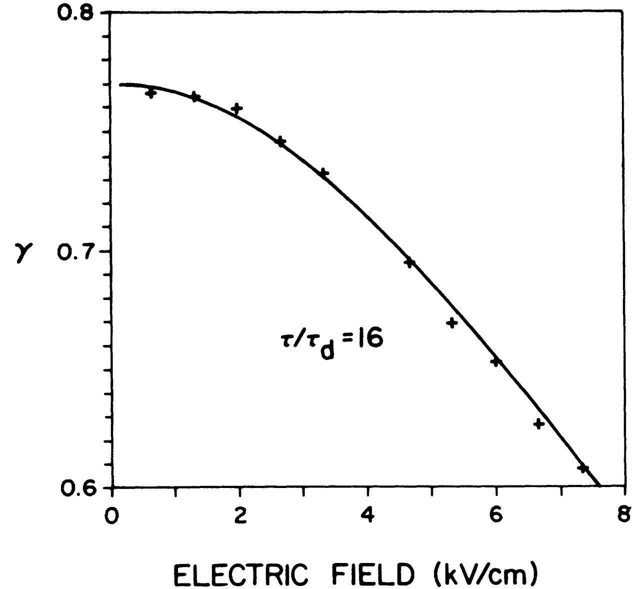

FIG. 6. The normalized conductivity grating amplitude [Eq. (37)] vs the applied electric field. Experimental results and a fit to the theory with the parameters $b=\mu_n/\mu_p=1.9$ and $c=\tau/\tau_d=16$.

We now use the measured value of the carrier lifetime to obtain a theoretical fit to the data. The photoconductivity at the same light intensity was 8.7×10^{-6} $(\Omega\,\mathrm{cm})^{-1}$, and the dielectric relaxation time is found from Eq. (32) taking $\kappa=11.8$ to equal 0.115 μsec. We thus obtain $c=\tau/\tau_d\approx16$. Using this value of c a fit can be found only for values of b ranging from unity to 1.9, and α/β ranging from unity to 1.6. We have verified again that the exact relationship between τ_n and τ_p does not significantly affect the results. The solid line in Fig. 6 is obtained by solving Eq. (33) for N_1 and P_1 with $b=1.9$ and $\alpha/\beta=1$, and the subsequent substitution of N_1 and P_1 in Eq. (36). It is seen that an excellent fit is obtained.

We thus conclude that the ambipolar mobility in our sample is small, and that the electric field does separate the electrons from the holes. This result differs from our original interpretation of the experiment in Ref. 4 where local charge neutrality was assumed, and as a result too large a value of the ambipolar mobility was estimated. To check the consistency of our results the value of $b=\mu_n/\mu_p$ can also be independently obtained from Eq. (23) which assumes *a priori* that charge neutrality prevails. Since the diffusion length at the same light intensity was found to equal (0.15 μm)$\pm$5%, and the small-signal photoconductivity was 1.1×10^{-7} $(\Omega\,\mathrm{cm})^{-1}$ for $\Delta G=4\times10^{20}$ pairs/cm^3, we again obtain that b is close to unity. The use of Eq. (23) is justified since it is evident from Fig. 2 that for $c=16$ and for small values of b the measured diffusion length is very close to the value of the ambipolar diffusion length.

A ratio of the electron and hole drift mobilities of order unity is surprising since time-of-flight experiments usually yield a larger drift mobility of electrons by about a factor of 10.[25,26] A possible source of difference between the two types of measurements may be that time-of-flight measurement are carried out under transient conditions while the experiments reported here were performed under steady-state conditions. Quite possibly, μ_n/μ_p may be quite different in these two cases. Clearly, more experiments on other samples are needed to establish whether there are consistent differences between the drift mobility values obtained from time-of-flight measurements and the methods discussed here and to what extent such differences are sample dependent.

VI. DISCUSSION

The primary aim of this paper has been to examine the transport and recombination behavior of photocarriers in a semiconductor when they are generated sinusoidally along a given sample direction, as in a SSPG. While the simple case in which local neutrality was assumed has been discussed before,[1-4] the more general case where space-charge effects are taken into account is presented here for the first time. It is found that such space-charge effects are always present when the experiments are carried out at high electric fields unless the dielectric relaxation time is much shorter than the lifetime so that space charges are screened out. At low electric fields, space-charge effects are important only if the drift mobilities of electrons and holes are different. In this case too, they are screened out if the dielectric relaxation time is sufficiently short.

An understanding of these effects is necessary in order to ensure that SSPG measurements, which are normally carried out at low fields, yield the true ambipolar diffusion length L. We show that, strictly speaking, the measurements must be carried out as a function of electric field and the lifetime of the excess carriers must be known in order to calculate L from experimental data.

The theory described in this paper is quite general, i.e., it applies equally well to crystalline or amorphous semiconductors. Our emphasis, however, has been on amorphous materials since most of the experimental results using the SSPG technique have related to hydrogenated amorphous silicon.[1-4] In the amorphous case the transport equations are more complicated than for the crystalline case since most of the photocarriers are probably trapped in localized states. We have taken this point into account in formulating the transport equations and have also restricted the theory to the small-signal regime in which only the behavior of an incremental concentration of photocarriers is considered. Hence, a strong bias illumination is assumed so that the various parameters which enter the transport equation, namely the drift mobilities, the diffusion constants, and the lifetime, are defined with respect to the background carrier concentration produced by the bias light. Since all these parameters are likely to depend on the light intensity it would be very instructive to carry out such measurements for different light biases.

For a representative sample of hydrogenated amorphous silicon produced by glow discharge decomposition of silane we found that at a bias illumination of approximately 1 sun the lifetime of the total excess carriers, as distinct from that of only the free carriers, is about 2 μsec and that the drift mobilities of electrons and holes both approximately equal 10^{-2} cm^2/V sec. Since the mobility ratio is of the order of magnitude unity and the ratio of lifetime to dielectric relaxation time is 16 for this sample, the low-field SSPG determination of an ambipolar diffusion length of 0.15 μm can be trusted. It would clearly be desirable to carry out such measurements on a number of samples in order to ascertain to what extent the values found in this work are representative.

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APPENDIX: THE SMALL-SIGNAL PHOTOCURRENT DECAY EXPERIMENT

In the small-signal photocurrent decay experiment the generation rate is abruptly decreased from a value $G+\Delta G$ to G where $\Delta G \ll G$, and the subsequent small change in the photocurrent is measured as a function of time. This experiment is similar to that analyzed by Pandya and Schiff,[27] with the distinction that in the initial stage of the decay in our case the carriers are in thermal equilibrium. As outlined in Ref. 27 two possible regimes have to be considered. In the first regime, which is called "A trapping," the carrier detrapping (trapping) times are shorter than the recombination time. In the second regime, which is called "B trapping," the opposite relation between trapping and recombination time exists. In the A-trapping regime the carriers are thermalized during the decay so that the photocarrier drift mobility is nondispersive and constant. As a result the decay can be described by a single exponential with a time constant which is the photocurrent response time as given by Rose.[15] Using the formalism of this paper we obtain

$$\frac{d}{dt}\Delta\sigma=(\mu_n'+\mu_p')q\frac{d}{dt}\Delta N \ , \tag{A1}$$

and

$$\frac{d}{dt}\Delta N=\Delta R=\frac{\Delta N}{\tau} \ , \tag{A2}$$

thus the time constant of the decay is seen to be the carrier lifetime as defined in Eqs. (13) and (14).

In the B-trapping regime, on the other hand, since the detrapping time of the carriers is longer than the recombination time, the photocarrier drift mobility is no longer constant during the decay. In this case Eq. (A1) is not valid due to the time dependence of the drift mobilities, and the decay is nonexponential. As pointed out in Ref.

27 A trapping is expected at high bias illumination, and B trapping at low bias illumination. This was indeed observed in our experiments. The decays at bias light intensities above about 50 mW/cm^2 were exponential whereas the decays at lower bias light intensities were nonexponential. A computer simulation of the decays has been carrier out by us and will be published elsewhere.

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