Theory of the steady-state-photocarrier-grating technique for obtaining accurate diffusion-length measurements in amorphous silicon

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This paper presents a theory that is the basis of the steady-state-photocarrier-grating (SSPG) technique, as a means of determining the diffusion length of photocarriers in amorphous semiconductors. Solving the SSPG transport problem, including the formulation of small-signal photocurrent, by a second-order perturbation approach reveals deficiencies in the existing SSPG theory, which is based on first-order perturbation theory. It is also shown that the SSPG data analysis done routinely, assuming *a priori* that local space-charge neutrality prevails, yields a severe overestimate of the diffusion length when the condition is not met experimentally. An extension of the theory to measure accurately the diffusion length by removing these defects inherent in the original SSPG formulation is demonstrated by its successful application to hydrogenated amorphous silicon. The experiments carried out at various illumination levels show that the correct value of the diffusion length and its light-intensity dependence indeed differ to a significant degree from the results obtained with use of the previous method. A physical interpretation of the measured intensity dependence is also given, assuming a trap-controlled photocarrier transport model.

I. INTRODUCTION

Following the growing interest in hydrogenated amorphous silicon (*a*-Si:H) and its alloy materials for solar-cell applications, the steady-state-photocarrier-grating (SSPG) technique has been introduced by Ritter, Zeldov, and Weiser as a simple and convenient way to measure the ambipolar diffusion length.[1] The ambipolar diffusion length is determined by less mobile or minority carriers, which play a crucial role in determining the performance of photovoltaic cells, and is, in general, hardly assessed as compared with the transport parameters relevant to more mobile or majority carriers. The recent popularity of the SSPG technique among researchers of these materials[2–10] is greatly due to its experimental ease.

In the SSPG experiment, the sample is illuminated by two coherent light beams of different intensities which interfere and form a small grating superimposed on a large uniform illumination. When the grating spacing is much longer than the carrier diffusion length, a well-defined concentration grating of photocarriers is created in the sample. On the other hand, when the grating spacing is comparable to or shorter than the diffusion length an almost uniform carrier concentration occurs in spite of the nonuniform photogeneration. In order to detect the photocarrier grating amplitude that is a good indicator of the diffusion length, a photocurrent perpendicular to the grating fringes is measured in this technique. The SSPG theory developed by Ritter, Zeldov, and Weiser on the basis of a small-signal approach connects the magnitude of the measured photocurrent with the ambipolar diffusion length under the assumption of local space-charge neutrality.[1,2] The consequences derived from their theory are at present widely utilized for the analysis of SSPG experimental data.

We wish here to discuss the validity of prevailing SSPG theory in detail, to propose an extended technique for determining the diffusion length beyond the charge-neutrality restriction, as well as to present experimental results for *a*-Si:H obtained at various illumination levels. In Sec. II we first attack the SSPG transport problem, in which a small incremental concentration of photocarriers is mathematically treated as a perturbation to the background concentration, and show that a complete solution for resulting SSPG current cannot be gained from the previous theory starting with the first-order perturbation.[1–3] The SSPG theory is thereby corrected precisely taking into account the effects associated with the higher-order perturbation. It turns out through the theoretical analysis that a correction for magnitude of the SSPG current is highly necessitated in the case when space-charge effects become significant. In this theoretical section, we also emphasize the pitfalls of the original SSPG data analysis[1,2] which assume *a priori* that charge-neutrality prevails. To remove such an experimental uncertainty and to extend the applicable range of the diffusion length measurement, in Sec. III we introduce two independent experiments, the steady-state photoconductivity (SSPC) and frequency-resolved photocurrent (FRPC) experiments. Combining the SSPG data with the results of these measurements yields an accurate determination of the diffusion length which does not require charge-neutrality condition. We present the experimental data for *a*-Si:H obtained as a function of illumination level, from which it is proved that the previous SSPG diffusion-length measurements are erroneous to a significant degree in the conventional light intensity range 1–100 mW/cm^2.[1–8] A physical interpretation on the light-intensity dependence of measured diffusion length is also given, assuming the trap-controlled photo-

carrier transport model. In Sec. IV we summarize the main conclusions of this paper.

II. THEORETICAL ANALYSIS OF THE STEADY-STATE PHOTOCARRIER GRATING (SSPG) TECHNIQUE

A. Perturbation expansion for the transport equations

On the basis of the perturbation expansion theory, this section presents a thorough treatment manner of steady-state transport equations in the small-signal case. Such an adaptation of the small-signal approach has been found in some previous papers, in which their theoretical concerns are concentrated on the first-order perturbation in order to avoid a probable nonlinear nature of the transport equations.[3,11,12] As will be stressed in Sec. II B, the small-signal photocurrent in the SSPG experiment is governed by the nonlinear effects relevant to the higher-order correction terms involved in the perturbation expansion, whereas these effects have never yet been taken into account precisely. The clarification of the small-signal treatment with incorporation of the higher-order corrections is therefore an important prerequisite for placing the SSPG theory on a self-consistent basis.

In order to make this paper self-contained, we shall start with the formalism of transport equations. It may be convenient here to use the concept of drift mobility and effective diffusion constant[2,3] by which the transport equations can be represented in a simple fashion. The set of drift mobility and effective diffusion constant for electrons is defined by

$$\mu_n(N)=\mu_n^f N^f/N \quad \text{and} \quad D_n'(N)=D_n^f dN^f/dN \ ,$$

and that for holes is

$$\mu_p(P)=\mu_p^f P^f/P \quad \text{and} \quad D_p'(P)=D_p^f dP^f/dP \ .$$

In the above relations, N is the total electron concentration consisting of the free (N^f) and trapped (N^t) concentrations ($N=N^f+N^t$), and equivalently P ($=P^f+P^t$) the total hole concentration. $\mu_{n,p}^f$ and $D_{n,p}^f$ denote the band mobilities and band diffusion constants, respectively, which are the microscopic transport parameters assigned to free carriers. It is evident from the definitions that a standard Einstein relationship as found between the band mobilities and band diffusion constants; $D_{n,p}^f=(k_B T/q)\mu_{n,p}^f$, where k_B is Boltzmann's constant and T the temperature, is not maintained in the parameter counterparts introduced here.

The one-dimensional electron and hole conduction currents are expressed with these transport parameters by

$$J_n=q\mu_n(N)NE+qD_n'(N)dN/dx \ , \tag{1a}$$

$$J_p=q\mu_p(P)PE-qD_p'(P)dP/dx \ , \tag{1b}$$

where x is the one-dimensional spatial variable, and q the elementary electric charge. The space charge $q(P-N)$ is related to the electric field E by the Poisson equation $dE/dx=(q/\epsilon)(P-N)$, where ϵ denotes the dielectric constant. The steady-state continuity equations for the electron and hole currents under light illumination are represented by

$$G-R+(1/q)dJ_n/dx=0 \ , \tag{2a}$$

$$G-R-(1/q)dJ_p/dx=0 \ . \tag{2b}$$

Here G and R are the photogeneration rate and the recombination rate, respectively. The recombination rate is, in general, expressed as some function of electron and hole concentrations, $R=R(N,P)$.

In the small-signal approach, one considers that a weak nonuniform generation $G_1(x)$ is superimposed on a relatively strong uniform generation G_0. The generation rate is thus composed of the two parts:

$$G(x)=G_0+G_1(x) \ . \tag{3}$$

In the weak generation limit $G_1 \ll G_0$, the carrier concentrations N and P are expressed in the form of perturbation expansion

$$N(x)=N_0+N_1(x)+N_2(x)+\cdots \ , \tag{4a}$$

$$P(x)=P_0+P_1(x)+P_2(x)+\cdots \ . \tag{4b}$$

The subscripts $0,1,2,\ldots$ refer to the order of perturbation. In the following, we impose the condition $N_0=P_0$ assuming that no local net space charge is present due to the uniform generation G_0. This constitutes a simplified but acceptable physical picture if the material is homogeneous. The conduction currents J_n and J_p can be written as

$$J_n(x)=J_{n0}+J_{n1}(x)+J_{n2}(x)+\cdots \ , \tag{5a}$$

$$J_p(x)=J_{p0}+J_{p1}(x)+J_{p2}(x)+\cdots \ . \tag{5b}$$

The expression for each correction term in the above expansion is obtained from a couple of perturbation expansions for drift and diffusion terms contained in Eq. (1).

By using the expansion formula of carrier concentration (4), one can expand the drift and diffusion terms. The drift-related term for electrons is represented in this manner by

$$\mu_n(N)N=\mu_n N_0+\mu_n' N_1+\mu_n' N_2+\mu_n'' N_1^2/N_0+\cdots \ , \tag{6}$$

where the drift mobility μ_n on the right-hand side of the equation is

$$\mu_n=\mu_n(N_0)=\mu_n^f(N^f/N)_0 \ ,$$

and the small-signal drift mobility of the first order μ_n' and that of the second order μ_n'' are, respectively, defined by

$$\mu_n'=\mu_n^f(dN^f/dN)_0 \ ,$$

$$\mu_n''=\mu_n^f(N_0/2)(d^2N^f/dN^2)_0 \ .$$

In the above definitions, $(\ )_0$ means the value of $(\ )$ in the unperturbed case. The expansion of diffusion term is given for electrons by

$$D_n'(N)dN/dx = D_n'dN_1/dx + D_n'dN_2/dx + D_n''d(N_1^2/N_0)/dx + \cdots . \quad (7)$$

Here, lack of the zeroth-order term is due to the spatial uniformity of the unperturbed part of concentration. The effective diffusion constants in the small-signal case are defined by

$$D_n' = D_n'(N_0) = D_n^f(dN^f/dN)_0 ,$$

$$D_n'' = D_n^f(N_0/2)(d^2N^f/dN^2)_0 .$$

The corresponding expressions for holes are readily obtained by replacing N (and suffix n) with P (and suffix p). It is worthwhile stating here the standard Einstein relationship holds for the small-signal parameter-pairs; $\mu_{n,p}'$ and $D_{n,p}'$, $\mu_{n,p}''$ and $D_{n,p}''$. The relation has been shown by Ritter, Zeldov, and Weiser.[3] In a similar way, the recombination rate is also written in the form of perturbation series:

$$R(N,P) = N_0/\tau + N_1/\tau_n' + P_1/\tau_p' + N_2/\tau_n' + P_2/\tau_p' + (N_1^2/N_0)/\tau_n'' + 2(N_1P_1/N_0)/\tau_{np}'' + (P_1^2/P_0)/\tau_p'' + \cdots , \quad (8)$$

where the lifetime parameters are defined by

$$1/\tau = (R/N)_0 ,$$

$$1/\tau_n' = (dR/dN)_0 , \quad 1/\tau_p' = (dR/dP)_0 ,$$

$$1/\tau_n'' = (N_0/2)(d^2R/dN^2)_0 ,$$

$$1/\tau_{np}'' = (N_0/2)(d^2R/dN\,dP)_0 ,$$

and

$$1/\tau_p'' = (N_0/2)(d^2R/dP^2)_0 .$$

It should be noted that all the values of the transport parameters introduced above are determined through the background carrier concentrations $N_0 = P_0$ generated by G_0.

Using Eqs. (1)–(8), one consequently obtains the transport equations in the small-signal case, which are classified by the order of perturbation as follows. The equations for unperturbed parts are simply given by

$$J_{n0} = q\mu_n N_0 E_0 , \quad (9a)$$

$$J_{p0} = q\mu_p P_0 E_0 , \quad (9b)$$

$$N_0 = P_0 = G_0\tau . \quad (9c)$$

Here, E_0 is an externally applied electric field, and is uniform in space within the context of the assumption that $N_0 = P_0$. The first-order carrier concentrations N_1 and P_1 are governed by the differential equations

$$J_{n1} = q\mu_n N_0 E_1 + q\mu_n' N_1 E_0 + qD_n' dN_1/dx , \quad (10a)$$

$$J_{p1} = q\mu_p P_0 E_1 + q\mu_p' P_1 E_0 - qD_p' dP_1/dx , \quad (10b)$$

$$G_1 - N_1/\tau_n' - P_1/\tau_p' + (1/q)dJ_{n1}/dx = 0 , \quad (10c)$$

$$G_1 - N_1/\tau_n' - P_1/\tau_p' - (1/q)dJ_{p1}/dx = 0 , \quad (10d)$$

$$dE_1/dx = (q/\epsilon)(P_1 - N_1) . \quad (10e)$$

A linearization found in the set of equations is a natural consequence of collection of the first-order terms, and enables us to easily obtain an analytical solution of the equations. The second-order equations, which contain nonlinear product terms such as N_1E_1 and N_1^2, are found to be

$$J_{n2} = q\mu_n N_0 E_2 + q\mu_n' N_1 E_1 + q(\mu_n' N_2 + \mu_n'' N_1^2/N_0)E_0 + q[D_n' dN_2/dx + D_n'' d(N_1^2/N_0)/dx] , \quad (11a)$$

$$J_{p2} = q\mu_p P_0 E_2 + q\mu_p' P_1 E_1 + q(\mu_p' P_2 + \mu_p'' P_1^2/P_0)E_0 - q[D_p' dP_2/dx + D_p'' d(P_1^2/P_0)/dx] , \quad (11b)$$

$$-N_2/\tau_n' - P_2/\tau_p' - (N_1^2/\tau_n'' + 2N_1P_1/\tau_{np}'' + P_1^2/\tau_p'')/N_0 + (1/q)dJ_{n2}/dx = 0 , \quad (11c)$$

$$-N_2/\tau_n' - P_2/\tau_p' - (N_1^2/\tau_n'' + 2N_1P_1/\tau_{np}'' + P_1^2/\tau_p'')/N_0 - (1/q)dJ_{p2}/dx = 0 , \quad (11d)$$

$$dE_2/dx = (q/\epsilon)(P_2 - N_2) . \quad (11e)$$

B. Photocurrent due to a weak illumination grating

The steady-state current continuity stating that the total conduction current $J = J_n(x) + J_p(x)$ must be constant at all x, makes it simple to derive the representation for the SSPG current. Before going into the practical description, we shall first give a generalized manner to treat the small-signal photocurrent produced by the illumination with a cyclic boundary condition $G_1(x) = G_1(x + \Lambda)$, which is encountered in the SSPG experiment. The current continuity permits us to write the expansion of J as

$$J = \langle J_0 \rangle + \langle J_1 \rangle + \langle J_2 \rangle + \cdots . \quad (12)$$

Here, $\langle\ \rangle$ expresses the spatial average defined by the integration from $x = 0$ to Λ divided by Λ. It is readily found that contributions of all the diffusion terms vanish in the spatial average process due to the periodicity, so that the average currents $\langle J_0 \rangle$, $\langle J_1 \rangle$, $\langle J_2 \rangle$, etc. are comprised solely of the drift terms. Averages of the electric-field corrections $\langle E_1 \rangle$, $\langle E_2 \rangle$, etc. are also equal to zero as is easily shown from the sum rule that $\langle E \rangle = \langle E_0 \rangle + \langle E_1 \rangle + \langle E_2 \rangle + \cdots = E_0$. The average currents are thus formulated by

$$\langle J_0 \rangle = \sigma_0 E_0 , \quad (13a)$$

$$\langle J_1 \rangle = \langle \sigma_1 \rangle E_0 , \quad (13b)$$

$$\langle J_2 \rangle = \langle \sigma_1 E_1 \rangle + \langle \sigma_2 \rangle E_0 , \quad (13c)$$

where the conductivities σ_0, σ_1, and σ_2 are, respectively, given by

$$\sigma_0 = q(\mu_n + \mu_p)N_0 ,$$

$$\sigma_1 = q(\mu_n' N_1 + \mu_p' P_1) ,$$

$$\sigma_2 = q(\mu_n' N_2 + \mu_p' P_2 + \mu_n'' N_1^2/N_0 + \mu_p'' P_1^2/P_0) .$$

At this level of our theory, the average concentrations $\langle N_1 \rangle$ and $\langle P_1 \rangle$ can be derived directly from the equations obtained by averages on both sides of Eqs. (10c)–(10e). A solution of the equations is found to be

$$\langle N_1 \rangle = \langle P_1 \rangle = \langle G_1 \rangle \tau' , \tag{14}$$

where τ' denotes a common small-signal lifetime defined by

$$1/\tau' = 1/\tau_n' + 1/\tau_p' .$$

What is read from combining Eq. (13b) with (14) is that any oscillating components of the light illumination do not contribute to the first-order average current. In this sense, the SSPG theory remains incomplete without the precise inclusion of effects of the second-order perturbation. The similar average procedure for Eqs. (11c)–(11e) allows us to obtain the reduced expressions for the second-order average concentrations $\langle N_2 \rangle$ and $\langle P_2 \rangle$, which are contained in the expression of $\langle \sigma_2 \rangle$, yielding

$$\langle N_2 \rangle = \langle P_2 \rangle = -(\tau'/N_0)(\langle N_1^2 \rangle/\tau_n'' + 2\langle N_1 P_1 \rangle/\tau_{np}'' + \langle P_1^2 \rangle/\tau_p'') . \tag{15}$$

From Eqs. (13c) and (15), it is found that the second-order average current can be determined from solving the first-order transport equation (10) and computing the averages of product terms: $\langle N_1 E_1 \rangle$, $\langle P_1 E_1 \rangle$, $\langle N_1^2 \rangle$, $\langle N_1 P_1 \rangle$, and $\langle P_1^2 \rangle$.

In order to proceed with our theoretical consideration into the SSPG current, here we will briefly outline the SSPG experimental setup.[1,2] In the experiment, the sample is illuminated uniformly by a laser beam of intensity I_0, which sets the value of generation rate G_0. A small portion of the beam I_1 is split off, passes through a light chopper, and impinges on the sample at an angle θ with I_0. Care is taken to ensure that the chopping frequency is low enough for the steady-state approximation to be valid. The ac photocurrent is measured under the two conditions that two beams have parallel polarization and those have perpendicular polarization. In the former case, the interference of beams produces an illumination grating with the period $\Lambda = (\lambda/2)\sin(\theta/2)$, where λ is the light wavelength. The perturbation to generation rate G_0 is then written by

$$G_1(x) = g_0 + g_K \cos(Kx) \quad \text{with } K = 2\pi/\Lambda , \tag{16}$$

where $G_0 : g_0 : g_K = I_0 : I_1 : 2\gamma_0 (I_0 I_1)^{1/2}$, and γ_0 $(0 < \gamma_0 \leq 1)$ is an interference quality factor.[1,2] When two beams have perpendicular polarization and thus do not interfere, G_1 is equal to g_0.

The sinusodial form of the generation rate greatly simplifies the solution of transport equations so far as those classified into the first-order is concerned.[3] The first-order carrier concentration is also composed of a small sinusoidal part superimposed on a uniform concentration:

$$N_1(x) = n_0 + \mathrm{Re}[\tilde{n}_K \exp(iKx)] , \tag{17a}$$

$$P_1(x) = p_0 + \mathrm{Re}[\tilde{p}_K \exp(iKx)] . \tag{17b}$$

Applying Eq. (14) readily yields $n_0 = p_0 = g_0 \tau'$, which corresponds to the solution in the case of no interference. $\tilde{n}_K$ and $\tilde{p}_K$ are complex numbers that give the amplitudes and phases of sinusoidal part. Substituting the above expressions into Eq. (10), one obtains two coupled linear equations for the unknown concentrations $\tilde{n}_K$ and $\tilde{p}_K$:

$$g_K \tau' = \tilde{n}_K [\delta + bc/(b+1) - iKL_{en} + K^2 L_{dn}^2] + \tilde{p}_K [\eta - bc/(b+1)] , \tag{18a}$$

$$g_K \tau' = \tilde{n}_K [\delta - c/(b+1)] + \tilde{p}_K [\eta + c/(b+1) + iKL_{ep} + K^2 L_{dp}^2] , \tag{18b}$$

where $L_{en,p} = \mu_{n,p}' E_0 \tau'$ are the drift lengths and $L_{dn,p} = (D_{n,p}' \tau')^{1/2}$ are the diffusion lengths.[13] The dimensionless constants in the equations are given by[3]

$$b = \mu_n/\mu_p , \quad c = \tau'/\tau_{\mathrm{diel}} ,$$

and

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

τ_{diel} denotes the dielectric relaxation time defined with the background conductivity σ_0 by $\tau_{\mathrm{diel}} = \epsilon/\sigma_0$.

For the sake of convenience, here we introduce another dimensionless constant r defined by[3]

$$br = \mu_n'/\mu_p' = L_{en}/L_{ep} = L_{dn}^2/L_{dp}^2 .$$

Using the definitions for the drift mobilities $\mu_{n,p}$ and the small-signal drift mobilities $\mu_{n,p}'$, one can write the expression for r by the ratio of the derivatives

$$r = [d \ln(P)/d\varepsilon_{Fp}]_0 / [d \ln(N)/d\varepsilon_{Fn}]_0 ,$$

where $\varepsilon_{Fn,p}$ denote the quasi-Fermi levels. The mathematical manipulation in deriving the above expression is justified when free carriers obey Boltzmann statistics. The derivatives on the right-hand side of the equation are tightly correlated with the energy distribution of trap states. Here it may be useful to give the expressions for the ratio r in two probable cases suggested for a-Si:H; a monotonic exponential distribution[14] and a structured distribution with a sharp drop at shallow energy position.[15,16] Assuming the exponential trap distribution, one has $r = T_c/T_v$ where $T_{c,v}$ are the characteristic temperatures describing the exponential distributions for electron and hole traps, respectively[3]. On the other hand, when the majority of trap states are shallower than quasi-Fermi-levels present, the total carrier concentration varies in proportion to the free-carrier concentration as will be discussed in Sec. III C, so that the expression for r is rewritten by replacing the characteristic temperature by an environment temperature.

Using Eqs. (13) and (18), the first-order SSPG current $\langle J_1 \rangle = \langle \sigma_1 \rangle E_0$ is readily found to be

$$\langle \sigma_1 \rangle E_0 = q(\mu_n' + \mu_p') n_0 E_0 \ . \tag{19}$$

This expression is valid whether the illumination grating is produced or not. The second-order SSPG current $\langle J_2 \rangle$ is given by the sum of $\langle \sigma_1 E_1 \rangle$ and $\langle \sigma_2 \rangle E_0$. Computing the averages of several product terms yields

$$\langle \sigma_1 E_1 \rangle = -(q^2/2\epsilon K)(\mu_n' + \mu_p')|\tilde{n}_K||\tilde{p}_K|\sin[\mathrm{Arg}(\tilde{n}_K) - \mathrm{Arg}(\tilde{p}_K)] \ , \tag{20a}$$

$$\langle \sigma_2 \rangle E_0 = (q/2N_0)E_0(\mu_n''|\tilde{n}_K|^2 + \mu_p''|\tilde{p}_K|^2 - (\mu_n' + \mu_p')\tau' \times \{|\tilde{n}_K|^2/\tau_n'' + 2|\tilde{n}_K||\tilde{p}_K|\cos[\mathrm{Arg}(\tilde{n}_K) - \mathrm{Arg}(\tilde{p}_K)]/\tau_{np}'' + |\tilde{p}_K|^2/\tau_p''\}) \ . \tag{20b}$$

In arriving at Eq. (20b), we neglect the terms containing the factor n_0^2. This neglect is verified when the weak excitation condition that $g_0^2/g_K^2 = I_1/4\gamma_0^2 I_0 \ll 1$ is met. In the case of no interference, both $|\tilde{n}_K|$ and $|\tilde{p}_K|$ are equal to zero so that within the approximation up to the second order the SSPG current is dominated solely by $\langle \sigma_1 \rangle E_0$.

Our expression for the SSPG current differs from those derived from the previous works.[3,10] Recently, Li's work[10] has concluded that the second-order SSPG current $\langle J_2 \rangle$ is given by $\langle q(\mu_n N_1 + \mu_p P_1)E_1 \rangle$, which corresponds to $\langle \sigma_1 E_1 \rangle$ with replacing $\mu_{n,p}'$ by $\mu_{n,p}$. The discrepancy arises from his simplified theory starting with the assumption that drift mobility and effective diffusion constant are concentration independent. As is evident, the model regarding these parameters as constant cannot deal with the trap-controlled photocarrier transport which would occur in the amorphous semiconductors with the localized states distributed within the band gap, and hence limits its applicability specifically to trap-free semiconductors. Taking into account the effects of traps results in introducing the small-signal transport parameters as well as the additional linear term $\langle \sigma_2 \rangle E_0$ as is in Eq. (20). The effects of traps are in a manner similar to ours considered in the SSPG theory originally derived by Ritter, Zeldov, and Weiser,[3] but a correct formulation of SSPG current is not achieved in their paper, as will be intensively discussed in the following part of this paper. The detailed discussion including comparison with our corrected theory is of great significance because their theory is widely used for analyzing the SSPG experimental data at present.

Finally we will give expressions for some important experimental parameters. Following Ritter, Zeldov, and Weiser we now define β (Refs. 1 and 2) as the ratio between the currents measured in the cases of interference and of no interference:

$$\beta = 1 + \langle \sigma_1 E_1 \rangle / \langle \sigma_1 \rangle E_0 + \langle \sigma_2 \rangle / \langle \sigma_1 \rangle \equiv 1 - 2\gamma_0^2 \alpha \Gamma \ . \tag{21}$$

Here, α is given by

$$\alpha = (\mu_n' + \mu_p')\tau'/(\mu_n + \mu_p)\tau \ . \tag{22}$$

It is easily understood that this parameter corresponds to the exponent in power-law light-intensity dependence of photoconductivity $\sigma \propto G^{\alpha}$. The SSPG parameter Γ is formulated using Eqs. (19) and (20) by

$$\Gamma = n_K p_K \sin\Phi/KL_{\mathrm{diel}} + [(\mu_n + \mu_p)/(\mu_n' + \mu_p')][\tau'(n_K^2/\tau_n'' + 2n_K p_K \cos\Phi/\tau_{np}'' + p_K^2/\tau_p'') - (\mu_n'' n_K^2 + \mu_p'' p_K^2)/(\mu_n' + \mu_p')] \ , \tag{23}$$

where $n_K = |\tilde{n}_K|/g_K\tau'$ and $p_K = |\tilde{p}_K|/g_K\tau'$ are the normalized amplitudes of electron and hole concentration gratings, $\Phi = \mathrm{Arg}(\tilde{n}_K) - \mathrm{Arg}(\tilde{p}_K)$ the phase shift between them, and $L_{\mathrm{diel}} = (\mu_n' + \mu_p')E_0\tau_{\mathrm{diel}}$ the dielectric relaxation length. The first and the second terms on the right-hand side of Eq. (23) are obtained from the computations for $\langle \sigma_1 E_1 \rangle / \langle \sigma_1 \rangle E_0$ and $\langle \sigma_2 \rangle / \langle \sigma_1 \rangle$, respectively. The expression for the SSPG parameter Γ is quite general and complete in the sense that it is not restricted to any specific transport model, and the effect of the second-order perturbation is accurately taken into account.

C. The SSPG parameter in the lifetime and relaxation-time regimes

Examining the SSPG parameter Γ for all the probable cases is somewhat troublesome since there are a large number of the transport parameters that modify its value. It is here appropriate to reduce the expression for Γ so as to match our model for photocarrier transport in a-Si:H derived in Sec. III C. This removes complication in the theoretical analysis and thereby makes the discussion straightforward.

In the following, we neglect the second term on the right-hand side of Eq. (23), which is a minor component of Γ. The validity of neglect will be assessed in Sec. III C. Γ is simply rewritten by

$$\Gamma = n_K p_K \sin\Phi/KL_{\mathrm{diel}} \ . \tag{24}$$

The expression for Γ is further accessed in the Appendix by employing an alternative to the present treatment for the SSPG current. It is clear there that the parameter Γ is comprised of two components relevant to the drift

current Γ_e and to the diffusion current Γ_d:

$$\Gamma=\Gamma_e-\Gamma_d \ . \tag{25}$$

These two components are given by

$$\Gamma_e=[(\mu_n')^2n_K^2+(\mu_p')^2p_K^2+2\mu_n'\mu_p'n_Kp_K\cos\Phi]/(\mu_n'+\mu_p')^2 \ , \tag{26a}$$

$$\Gamma_d=(2E_d/E_0)[\mu_n'\mu_p'/(\mu_n'+\mu_p')^2]n_Kp_K\sin\Phi \ , \tag{26b}$$

where $E_d=(k_BT/q)K$ is the so-called diffusion field. Γ_e is identical to that previously derived by Ritter *et al.*[17] They have, however, employed Γ_e as an approximation for Γ in their SSPG analysis. As will be evident below, this could be a possible cause to yield misinterpretation of the SSPG experimental data.

The parameters one can directly set in the SSPG experiment are the grating period Λ, the generation rate G_0, and the electric field E_0. Among these parameters, we will first bring the electric field E_0 into focus of consideration. As is easily expected from Eq. (18), an application of electric field causes the reduction of the amplitudes of electron and hole concentration grating as well as the phase slip between them. A criterion for a negligible electric field is readily found from comparing the drift terms $KL_{en,p}$ with the diffusion terms $K^2L_{dn,p}^2$ and is written by[9]

$$E_0 \ll E_d \ . \tag{27a}$$

This weak field condition indicates that the concentration grating is diffusion controlled. Even if the condition (27a) is not met, the effect of field still remains in a negligible range so far as the alternative conditions that $KL_{en} \ll bc/(b+1)$ and $KL_{ep} \ll c/(b+1)$ are both fulfilled. The corresponding criterion for $r=1$ is simply expressed by

$$KL_{\rm diel} \ll 1 \ . \tag{27b}$$

It might be worthwhile to mention that the inequality is rewritten as $2\pi(L_{en}+L_{ep})/\Lambda \ll \tau'/\tau_{\rm diel}$. This condition states that in the regime where the carrier lifetime is much longer than the dielectric relaxation time the field plays a negligible role in determining the state of concentration grating even when the drift length is comparable to or greater than the grating period. Thus Eq. (27) is intimately correlated with the transition between the lifetime and relaxation-time regimes in the presence of an externally applied field.[3] Detailed aspects in both regimes will be discussed below in conjunction with a low-field SSPG.

We next turn to the effect of generation rate G_0. Readily evident is that varying the illumination level directly modifies the value of dielectric relaxation time $\tau_{\rm diel}$. It must be also borne in mind through the consideration that the various parameters which enter the transport equations, such as the drift mobilities, the diffusion constants, and the lifetime, are defined with respect to the background carrier concentrations generated by G_0. In other words, the transport parameters are, in general, tuned by G_0. For the sake of simplicity, we here restrict our attention to the low-field case, which is justified by either Eq. (27a) or Eq. (27b). In this case, the concentration gratings of both types of carriers are in phase with the illumination grating. The expressions for normalized grating amplitudes n_K and p_K are reduced to

$$n_K=(1+K^2L_{dp}^2/c)/A_K \ , \tag{28a}$$

$$p_K=(1+K^2L_{dn}^2/c)/A_K \ , \tag{28b}$$

where the denominator A_K is given with the ambipolar diffusion length $L_{\rm amb}=[(\mu_nL_{dp}^2+\mu_pL_{dn}^2)/(\mu_n+\mu_p)]^{1/2}$ by

$$A_K=1+K^2L_{\rm amb}^2+[(K^2L_{dn}^2+\delta)(K^2L_{dp}^2+\eta)-\delta\eta]/c \ .$$

The expression for the SSPG parameter Γ is also simplified to

$$\Gamma=B_K/A_K^2 \ , \tag{29}$$

with

$$B_K=1+2K^2[L_{dn}^2L_{dp}^2/(L_{dn}^2+L_{dp}^2)]/c \ .$$

The drift-current component Γ_e and the diffusion-current component Γ_d are expressed by

$$\Gamma_e=B_K^2/A_K^2 \ , \tag{30a}$$

$$\Gamma_d=B_K(B_K-1)/A_K^2 \ . \tag{30b}$$

It is of importance to obtain the reduced expressions of grating amplitudes n_K and p_K as well as the SSPG parameter Γ for two extreme cases, the lifetime and relaxation-time regimes. The lifetime regime refers to the case where the dielectric relaxation time $\tau_{\rm diel}$ is much shorter than the carrier lifetime, and could be encountered at an appropriately strong background illumination. The above expressions are reduced in the $c\rightarrow\infty$ limit to

$$n_K=p_K=1/(1+K^2L_{\rm amb}^2) \ , \tag{31}$$

$$\Gamma=\Gamma_e=1/(1+K^2L_{\rm amb}^2)^2 \ . \tag{32}$$

Local charge neutrality and ambipolarity manifest themselves in the identical grating amplitude $n_K=p_K$. In this regime, the ambipolar diffusion length $L_{\rm amb}$ can be determined immediately from the Γ value measured in the SSPG experiment. We also note that the correction of the diffusion-current component is not necessary in this extreme case.[18] In the relaxation-time regime, the opposite relation stands between $\tau_{\rm diel}$ and τ'. Then, concentration grating is no longer simply controlled by the ambipolar diffusion, and the difference between diffusion lengths for electrons and holes produces space charge. We have the expressions in the $c\rightarrow 0$ limit

$$n_K=1/(\delta+br\eta+K^2L_{dn}^2) \ , \quad p_K=brn_K \ , \tag{33}$$

$$\Gamma=\Gamma_e-\Gamma_d=0 \ . \tag{34}$$

The magnitudes of Γ_e and Γ_d coincide with each other in the relaxation-time regime. This indicates that the SSPG theory given by Ritter *et al.*, which loses the correction of diffusion-current component Γ_d, completely breaks

when approaching the relaxation-time regime.

The numerically computed SSPG parameter Γ is displayed in the graphical form in Fig. 1. The parameter set assumed here is $r=1$, $\delta=\eta$, and $\Lambda=10L_{amb}$. This plot shows that when c becomes large Γ approaches that in the ambipolar case. On the other hand, when c becomes small Γ goes to zero. This tendency is in good accordance with the analytical result given above. Γ is found to deviate from Γ_e with going apart from the lifetime regime. The discrepancy is most pronounced at $b=1$ (equivalent with $br=1$ in this plot). The Γ_d correction is therefore highly necessary, especially in the small b (br) case. Since the assumptions $r=1$ and $\delta=\eta$ are not accepted in general, we must address the effects of varying them. It is easily understood that changing these values has a minor effect on the parameter Γ in the vicinity of the two extreme limits $c\to\infty$ and $c\to 0$. In the transition region between them, however, the effect becomes serious and cannot be neglected, as found in Fig. 2, where the c dependence of Γ is plotted for various values of r and $\delta(=1-\eta)$.

In the SSPG experiment, Γ is measured for several grating periods, and Eq. (32) is utilized for the L_{amb} determination. As learned above, the procedure will yield an erroneous estimation when the lifetime regime is not met practically. The calculated apparent diffusion length L_{app}, which is obtained from the $1/\sqrt{\Gamma}$ vs K^2 plot as normally done in the SSPG data analysis,[5,6] is plotted in Fig. 3 in the form of the ratio L_{app}/L_{amb}. We again set $r=1$ and $\delta=\eta$ in the calculation. The linearity in the $1/\sqrt{\Gamma}$ vs K^2 plot, where $K^2L_{amb}^2$ is varied from 0.1 to 1, is approximately sustained with changing the values of b and c in the present range of calculation. As also expected from the plot shown in Fig. 1, L_{app} is found to deviate from L_{amb} to a significant extent depending on b unless c

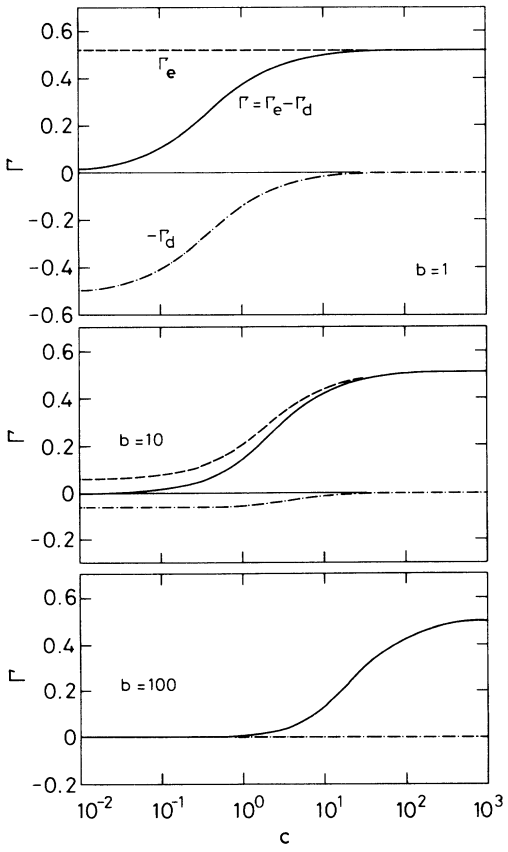

FIG. 1. Theoretical plot of the SSPG parameter $\Gamma=\Gamma_e-\Gamma_d$ vs $c=\tau'/\tau_{diel}$, for $\mu_n'/\mu_p'=\mu_n/\mu_p$ and $\tau_n'=\tau_p'$. The parameter $b=\mu_n/\mu_p$ is taken as 1, 10, and 100. The grating spacing Λ is chosen to equal ten times ambipolar diffusion length L_{amb}.

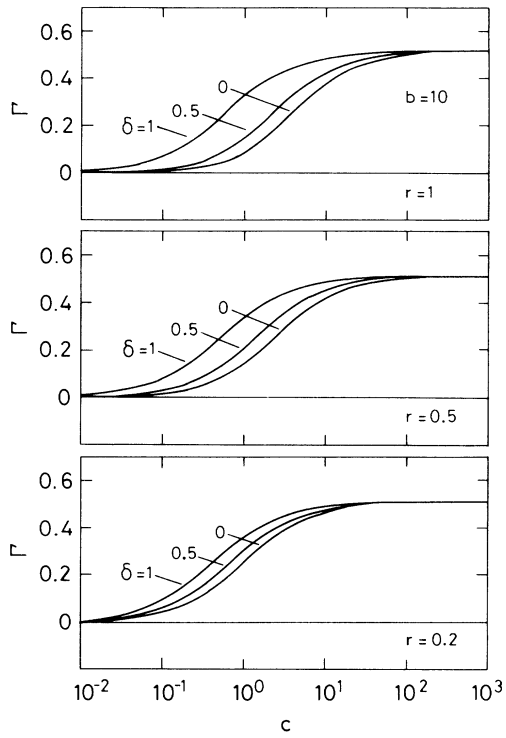

FIG. 2. Theoretical plot of the SSPG parameter Γ vs $c=\tau'/\tau_{diel}$, for $b=\mu_n/\mu_p=10$ and $\Lambda=10L_{amb}$. The parameters r and δ are the ratios $(\mu_n'/\mu_p')/b$ and τ'/τ_n', respectively.

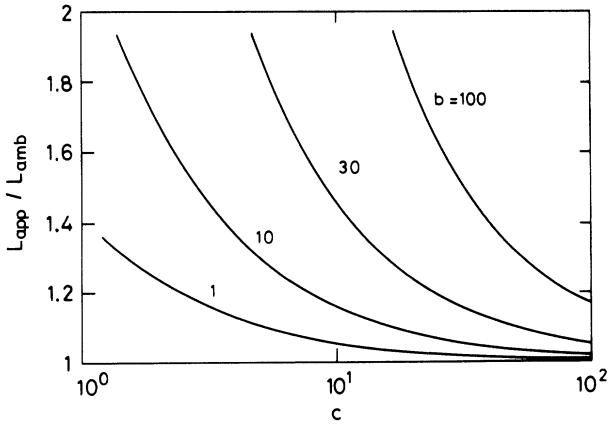

FIG. 3. Theoretical plot of the ratio L_{app}/L_{amb} vs $c=\tau'/\tau_{diel}$, for $\mu_n'/\mu_p'=\mu_n/\mu_p$ and $\tau_n'=\tau_p'$. L_{app} is the apparent diffusion length obtained from the $1/\sqrt{\Gamma}$ vs $K^2(=4\pi^2/\Lambda^2)$ plot, where $K^2L_{amb}^2$ is varied from 0.1 to 1. The parameter $b=\mu_n/\mu_p$ is taken as 1, 10, 30, and 100.

is sufficiently large.

We must stress here that it is impossible to draw any conclusion about L_{amb} only from the SSPG measurement when one does not know the values of b, c, r, and δ $(=1-\eta)$. In Sec. III, we will tackle the problem by using the SSPC and FRPC techniques, which determine the small-signal mobility-lifetime product $(\mu_n'+\mu_p')\tau'$ and the small-signal lifetime τ', respectively. Combining them with the SSPG experimental data enables us to evaluate the unknown parameters $b,c,\ldots$, as well as the diffusion lengths L_{amb} and $L_{dn,p}$.

III. DIFFUSION LENGTH MEASUREMENTS IN HYDROGENATED AMORPHOUS SILICON

A. Determination of the small-signal lifetime

As mentioned in Sec. II C, the ratio between the carrier lifetime τ' and the dielectric relaxation time τ_{diel} is one of the important parameters for applying the SSPG theory to the experimental data. The dielectric relaxation time τ_{diel} is directly determined from the background photoconductivity. On the other hand, the lifetime τ', which is equivalent with the response time of small-signal photoconductivity, can be measured from the photocurrent decay experiment as performed by Ritter, Zeldov, and Weiser.[3] Another possible way to measure the response time is the frequency domain study of the modulated photocurrent, which in this context refers to the ac component of photocurrent and its phase shift with respect to modulated optical illumination. An advantage when employing the frequency domain experiments is use of lock-in techniques with improved signal-to-noise ratios which allow us to accurately measure the small change in photocurrent of the present interest.

A 1-μm-thick-sample of a-Si:H deposited on a glass substrate was used in the experiments. The a-Si:H film was prepared by rf plasma decomposition from a $[SiH_4]/[H_2]=1:9$ gas mixture. The substrate temperature during decomposition was 250 °C. Contacts were the coplanar Al electrodes with a gap of 1 mm deposited on top of the sample. The sample was exposed for 4 h to a He-Ne laser (632.8 nm, 150 mW/cm^2) so as to minimize the effects of light-induced change in the transport parameters during measurements.

In the FRPC experiment, the sample was illuminated by a He-Ne laser beam of which intensity was modulated to a depth of 5% sinusoidally by an acousto-optic modulator. The voltage applied between the electrodes was 200 V. The modulated part of photocurrent was detected by a lock-in amplifier, and the phase lag ϕ was recorded as a function of the modulation frequency f. The phase lag ϕ is expressed with the small-signal lifetime τ' by

$$\tan(\phi)=2\pi f\tau' , \tag{35}$$

when assuming that the modulation frequency is low enough to be smaller than the thermal release rate of a trapped carrier at a quasi-Fermi-level.[19,20] Such a low-frequency approximation is expected to be valid at a higher background illumination because of the resulting high-energy position of the quasi-Fermi-level. In the opposite case, the trapping and detrapping processes associated with localized states above the quasi-Fermi-level no longer play a negligible role in determining the phase lag, so that the linearity between $\tan(\phi)$ and f as expected from Eq. (35) tends to be destroyed.[19] Shown in Fig. 4 is the frequency dependence of $\tan(\phi)$ measured at the background light intensity $I_0=77$ mW/cm^2. An excellent linearity between $\tan(\phi)$ and f is read from the figure, indicating the plausibility of the low-frequency approximation. A slope of the linear relation yields an estimate of the lifetime at 2.6 μsec. This is in reasonable agreement with the previous estimation from the time-resolved measurement that $\tau'=1.8$ μsec at $I_0=70$ mW/cm^2.[3]

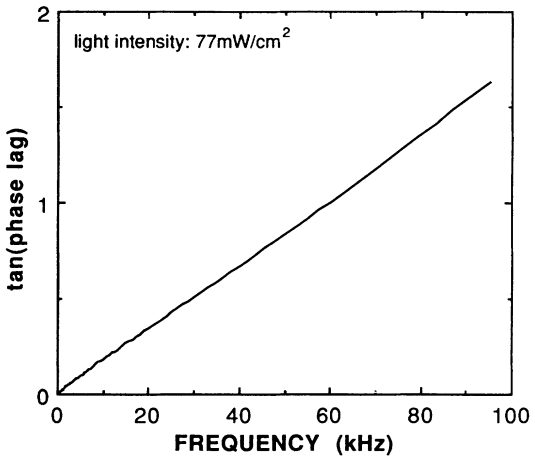

FIG. 4. Tangent of the phase lag of the small-signal photocurrent as a function of the modulation frequency, measured for the a-Si:H sample at the background light intensity of 77 mW/cm^2.

The lifetime data obtained at various light intensities are summarized in Fig. 5. The $\tan(\phi)$ spectrum measured at low illumination levels was nonlinear in the high-frequency region as predicted. In such a case, the estimation of lifetime τ' was performed for the linear range of spectrum seen in the lower frequency side. It is found in

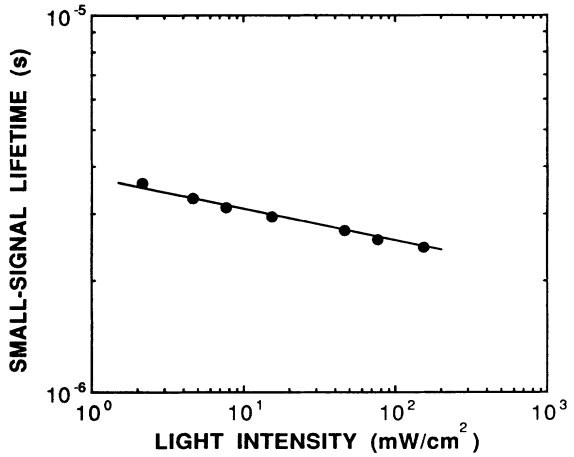

FIG. 5. Light-intensity dependence of the small-signal carrier lifetime τ' determined from the phase-shift data.

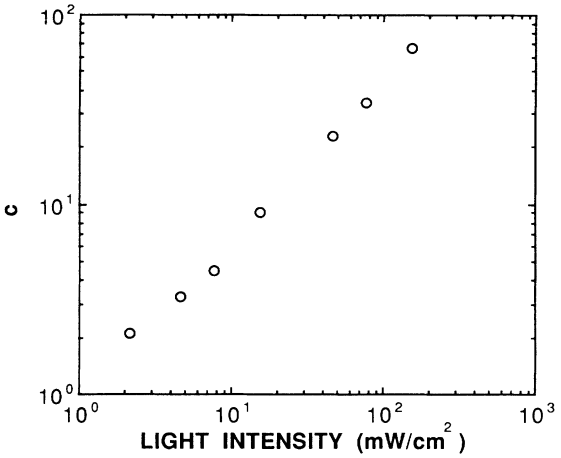

FIG. 6. Light-intensity dependence of the ratio $c=\tau'/\tau_{\text{diel}}$. τ_{diel} was calculated from the background photoconductivity with taking the dielectric constant as 12 for a-Si:H.

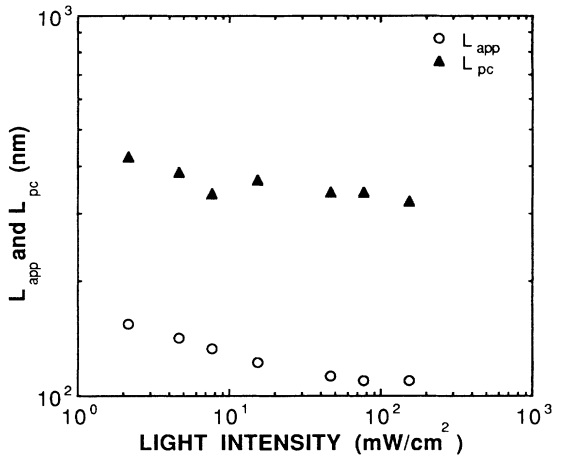

FIG. 7. Light-intensity dependence of L_{app} and L_{pc}, obtained from the SSPG and SSPC data, respectively.

Fig. 5 that the lifetime τ' decreases as $I_0^{-0.08}$. The weak intensity dependence implies that the dominant recombination process is monomolecular. The ratio $c=\tau'/\tau_{\text{diel}}$ is plotted in Fig. 6, and is seen to vary from 2.1 to 67 strongly depending on the illumination level.

B. Accurate evaluation of the diffusion length

This section gives the experimental results of SSPG and SSPC measurements. The outline of SSPG experiment is described in Sec. II B. A light from the He-Ne laser was used as the excitation light, and the intensity ratio I_1/I_0 was set at 5%. The chopping frequency was chosen at 40 Hz. At such a low frequency, the phase lag ϕ is as small as less than 1° in the range of illumination level studied, indicating that the steady-state approximation is practically applicable. The voltage applied between the contacts was 7 V ($E_0=70$ V/cm). The diffusion length L_{app} was determined from the SSPG experimental data obtained with varying the optical grating spacing from 0.6 to 2 μm. In this experimental selection for the applied voltage and the grating spacing, the external field E_0 is smaller than 10% of the diffusion field E_d, so that the weak-field condition (27a) is confirmed to be fulfilled. The SSPC experiments measure the small-signal photoconductivity in the case of uniform illumination, from which the mobility-lifetime product $(\mu_n'+\mu_p')\tau'$ is easily calculated in the usual manner.[20]

Figure 7 shows the measured diffusion length L_{app} as a function of the light intensity I_0. It is found in the figure that with increasing the light intensity from 2.1 to 150 mW/cm^2, L_{app} gradually decreases from 155 to 110 nm. This tendency agrees well with the previous SSPG experimental results.[5,7,8] The translation of L_{app} into L_{amb} is a subject of our central concern. The direct translation that $L_{\text{app}}=L_{\text{amb}}$, especially at a low illumination level where the ratio c is small, might yield a serious error as learned from Fig. 3. It would be therefore proper that the measured L_{app} is regarded only as the quantity given as function of the dimensionless constants b,c,r and $\delta(=1-\eta)$, and the ambipolar diffusion length L_{amb}.

The small-signal mobility-lifetime product obtained from the SSPC measurement is plotted in Fig. 7, in the form of $L_{pc}=[(k_BT/q)(\mu_n'+\mu_p')\tau']^{1/2}$. The mobility-lifetime product as well as L_{pc} are dominated by the more mobile or majority carrier while the ambipolar diffusion length L_{amb} is determined by the less mobile or minority carrier, and thus they are correlated by the drift mobility ratio. The relationship is given from their definitions by

$$L_{\text{amb}}^2/L_{pc}^2=b(r+1)/(b+1)(br+1)\ . \tag{36}$$

It is readily found that the unknown variable L_{amb} can be eliminated in the L_{app} calculation by using Eq. (36) with the measured L_{pc}.

We shall proceed to the diffusion length analysis for a moment with assumptions that $r=0.6$ and $\delta=1$, which enable us to obtain a simple theoretical plot of L_{app} only as a function of b, with using the measured values for c and L_{pc}. The former assumption is tentatively used in accordance with the conventional exponential-trap-distribution model ($T_c\cong 300$ K and $T_v\cong 500$ K in the representative sample of a-Si:H).[14] The detailed discussion on trap distribution will be given in Sec. III C, along with the physical interpretation on the experimental data obtained in this work. The assumption that $\delta=1$ means a larger hole lifetime τ_p' relative to the electron lifetime τ_n', and is naturally associated with what kind of recombination processes are operative. At present we do not have information enough to confirm physical ground of the assumption, whereas it will be shown below that this can be trusted experimentally.

Figure 8 displays the theoretical plot of L_{app} and L_{amb} obtained assuming $c=34.2$ and $L_{pc}=342$ nm estimated at $I_0=77$ mW/cm^2. According to SSPG measurements that $L_{\text{app}}=110$ nm, graphical solutions are readily found to be $b=30.5$ and $L_{\text{amb}}=96.9$ nm. The estimated b value can be used to deconvolute the ambipolar diffusion length L_{amb} into the electron and hole diffusion lengths $L_{dn,p}$. They are calculated to be $L_{dn}=333$ nm and $L_{dp}=77.8$ nm. At this illumination level, the c value is

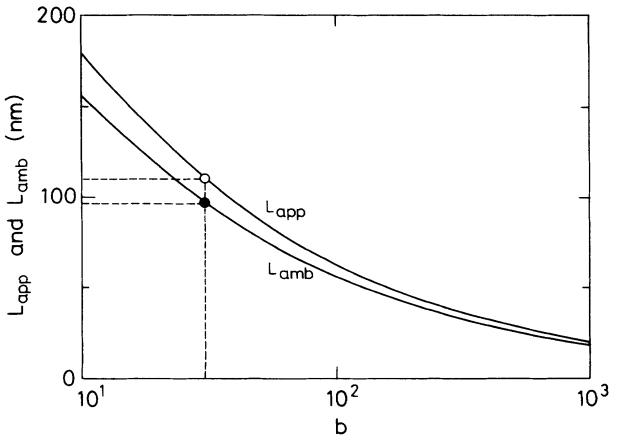

FIG. 8. Theoretical graph of L_{app} and L_{amb} vs the ratio $b=\mu_n/\mu_p$, for $c=34.2$ and $L_{pc}=342$ nm, which are the experimental results at the light intensity 77 mW/cm^2. The measured L_{app} value gives the solution for b and L_{amb}($b=34.5$ and $L_{amb}=97.3$ nm for $L_{app}=110$ nm).

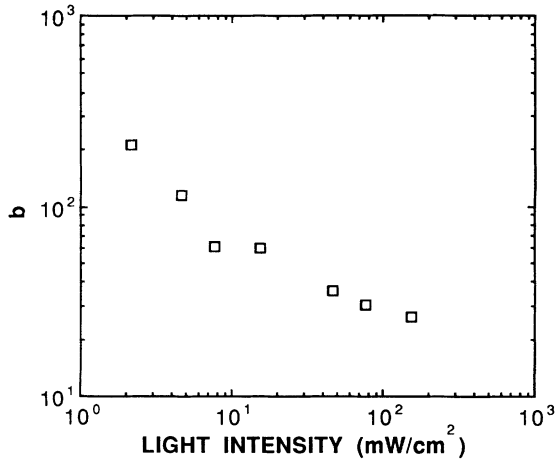

FIG. 9. Light-intensity dependence of the ratio $b=\mu_n/\mu_p$ obtained by the experimental analysis of the present work.

as large as compared to the b value, so that L_{app} does not so largely deviate from L_{amb}. The serious discrepancy is seen at lower illumination levels as expected. As an example, for $c=4.48$ and $L_{pc}=339$ nm estimated at $I_0=7.7$ mW/cm^2, we obtain $b=60.9$ and $L_{amb}=73.8$ nm for $L_{app}=133$ nm. In a similar way, $L_{dn,p}$ are determined to be 336 and 60.8 nm, respectively.

Here, we will briefly address the range of accuracy of present evaluation with the assumptions that $r=0.6$ and $\delta=1$. We have tested it by applying the r values ranging from 0.5 to unity and the δ values ranging from zero to unity to the determination of diffusion length. We confirmed that varying the r value does not significantly affect the result. Even maximum deviation in the estimation, which is found at the higher illumination level, is as small as 1% for the electron diffusion length and as 10% for the hole diffusion length. A change in the δ value has a more serious effect. When the δ value is set apart from unity, the deviation from the present estimation gets larger, and at lower illumination level it is not possible to find a solution for the diffusion length from the theoretical plot, indicating the physical inconsistency in used parameters. The fact that the solution is obtainable throughout the illumination level studied only when δ is close to unity lends a trust to the present evaluation, although the possibility that the δ value is varied with the light intensity could not be ruled out.

The estimated b value is plotted as a function of the light intensity I_0 in Fig. 9, where the b value is found to decrease from 213 to 26.4 with increasing the light intensity. The present estimation of drift mobility ratio larger than 10 serves a firm basis of the usual photoconductivity analysis in which a unipolar conduction due to electrons is traditionally assumed. On the contrary, Ritter, Zeldov, and Weiser have amounted the b value to be close to unity at $I_0=70$ mW/cm^2, using the relation (36) with the assumption that $L_{app}=L_{amb}$.[3] It can be read from their paper that the disagreement would be mainly ascribed to their incorrect estimation of the mobility-lifetime product done with ignoring correction for light reflection at sample surface. They have also measured the external electric-field dependence of the SSPG parameter Γ in the range higher than diffusion field, of which result was fit to theory to check the consistency of evaluation for b,c, and L_{amb}.[3] However, their approach would result in the misreading of data because of an application of the theory in which the diffusion-current component of Γ is not involved as pointed out in Sec. II C. In fact, our experimental result on the electric-field dependence is consistent with the corrected theory assuming the transport parameters determined here, and will be presented in our forthcoming publication with the detailed discussion.

Figure 10 presents the light-intensity dependence of the diffusion lengths L_{app}, L_{amb}, and $L_{dn,p}$. This figure shows an important conclusion reached by this work, namely, the measured diffusion length L_{app} indeed differs from the true ambipolar diffusion length L_{amb} to a significant degree. With a reduction of the light intensity, the ambipolar diffusion length L_{amb} decreases from

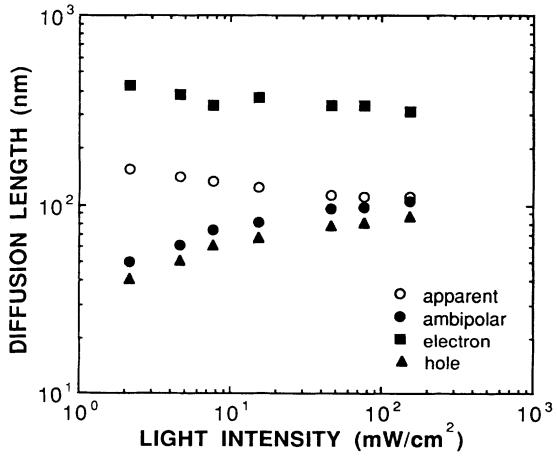

FIG. 10. Light-intensity dependence of the apparent and ambipolar diffusion lengths, and the electron and hole diffusion lengths.

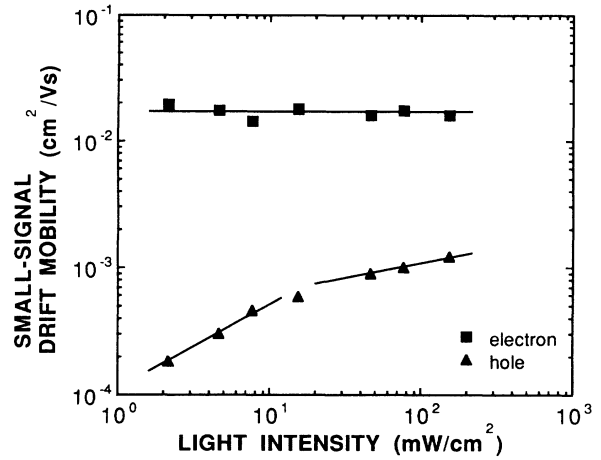

FIG. 11. Light-intensity dependence of the small-signal drift mobilities for electrons and holes.

104 to 50.2 nm whereas the apparent diffusion length L_{app} exhibits the opposite variation. Evidently, the deviation arises mainly from the decrease in the ratio $c=\tau'/\tau_{diel}$ as well as the increase in the ratio $b=\mu_n/\mu_p$, both of which tend to break the direct translation that $L_{app}=L_{amb}$. This result therefore contradicts a speculation made by Balberg that the SSPG experiments directly measure the correct value of L_{amb} and its light-intensity dependence in the range of conventional light intensities 1–100 mW/cm^2.[9]

Variation in $L_{dn,p}$ seen in Fig. 10 is due to that in the small-signal drift mobilities $\mu'_{n,p}$ multiplied by the carrier lifetime τ'. $\mu'_{n,p}$ decoupled from the products are plotted in Fig. 11. In this figure, it is clearly found that the electron mobility μ'_n is almost unchanged and kept at 1.7×10^{-2} cm^2/Vs while the hole mobility μ'_p increases from 1.85×10^{-4} cm^2/Vs to 1.21×10^{-3} cm^2/Vs with the light intensity. The striking feature in the curve for μ'_p is a switch of the exponent in the power-law intensity dependence from 0.65 to 0.25 around $I_0=10$ mW/cm^2. In Sec. III C, this point will be discussed in detail.

C. Physical interpretation on the light-intensity dependence of transport parameters

In this section, we give a physical interpretation of the present experimental data obtained as a function of the illumination level. As revealed from the measurements, the steady-state transport parameters vary depending on the light intensity. A possible source of this behavior would be that the trap-controlled transport of photocarriers is operative.

It is very instructive to first examine the relationship between the photoconductivity σ_0 and the lifetime τ'. The a-Si:H sample used in this work shows the power-law intensity dependence of photoconductivity with the exponent $\alpha=0.93$ in the illumination level studied. We can thereby obtain a phenomenological relation:

$$\mu_n N_0 \propto G_0^{\alpha} , \tag{37}$$

when taking into account the large difference between electron and hole drift mobilities confirmed experimentally. It can be accepted here that the electron drift mobility μ_n is intensity independent, being consistent with the fact that the small-signal drift mobility μ'_n is kept at constant. The constant electron drift mobility yields $R_0 \propto N_0^{1/\alpha}$, since in the steady state with uniform illumination $R=G$. We further assume that the above expression for recombination rate can approximately deal with the general case including nonuniform generation, namely,

$$R \propto N^{1/\alpha} . \tag{38}$$

Note that Eq. (38) is written without P so as to give $1/\tau'_p=0$ and to satisfy the assumption that $\delta=1$. Using Eqs. (37) and (38), the common small-signal lifetime τ' varies as

$$\tau' \propto G_0^{\alpha-1} . \tag{39}$$

As mentioned in Sec. III A, the exponent in the power-law intensity dependence of measured τ' is -0.08. The experimentally determined value is in excellent agreement with the value predicted from Eq. (39), $\alpha-1=-0.07$. This coincidence justifies the above description that addresses the photoconductivity and the recombination rate.

We next turn our focus to the light-intensity dependence of electron drift mobilities μ_n and μ'_n. The behaviors of μ_n and μ'_n are both governed by those of free and trapped electron concentrations. From a theoretical point of view, several approaches to assess them may be attempted. The most standard equations are the following:

$$N^f=N_c\exp[(\varepsilon_{Fn}-\varepsilon_c)/k_BT] , \tag{40}$$

$$N^t=\int d\varepsilon\, g_n(\varepsilon)\{1+\exp[(\varepsilon-\varepsilon_{Fn})/k_BT]\}^{-1} . \tag{41}$$

N_c is the effective density of states above the conduction-band edge ε_c, and $g_n(\varepsilon)$ the density of electron trap states. The Fermi-Dirac distribution with quasi-Fermi-level ε_{Fn} in Eq. (41) is applied as an approximation.[20]

Suppose that the majority of electron trap states are situated above the quasi-Fermi-level. This shallow trap assumption is qualitatively consistent with the electron trap distribution with a sharp drop suggested from the low-temperature drift-mobility measurements,[15,16] if the quasi-Fermi-level is moderately deep. Equation (41) can be then rewritten by

$$N^t \cong (N^f/N_c)\int d\varepsilon\, g_n(\varepsilon)\exp[(\varepsilon_c-\varepsilon)/k_BT] , \tag{42}$$

which indicates that N^t varies in linear proportion to N^f. Applying the relation to the definitions for drift mobilities, $\mu_n=\mu'_n$ being independent on N_0 as well as on G_0 is readily obtained. The constant drift mobility is in good agreement with the present experimental result. Another possible explanation for this data is that the electron trap states are distributed exponentially with T_c close to room temperature, as is widely used in model consideration of transport results on a-Si:H.[14] For $T_c=T$ we have

$$N^t \cong (N^f/N_c)(\varepsilon_c - \varepsilon_{Fn}) g_n(\varepsilon_c) \ . \quad (43)$$

As found from Eq. (43), this model also yields an approximate proportionality between N^f and N^t.

The behavior of hole drift mobility can be accounted for by assuming an exponential distribution of hole trap states: $g_p(\varepsilon) = g_p(\varepsilon_v)\exp[(\varepsilon_v - \varepsilon)/k_B T_v]$ where $T_v > T$. The trapped hole concentration then becomes

$$P^t \cong [k_B T_v g_p(\varepsilon_v)/\mathrm{sinc}(\pi T/T_v)](P^f/N_v)^{T/T_v} \ , \quad (44)$$

where N_v is the effective density of states for valence band, and ε_v the mobility edge. From Eq. (44) we readily obtain the light-intensity dependence of hole drift mobilities

$$\mu_p = (T/T_v)\mu_p' \propto P_0^{(T_v - T)/T} \propto G_0^{\alpha(T_v - T)/T} \ , \quad (45)$$

for $P^t \gg P^f$. The power-law intensity dependence is indeed found in the experimental result displayed in Fig. 11. The change in the exponent seen in the measured μ_p' curve is, in the context of this model, directly ascribed to the change in the slope of the exponential distribution. From the experimentally determined values for $\alpha(T_v - T)/T$, the characteristic temperatures are estimated to be 370 and 500 K, both of which describe the exponential distribution in a shallower energy side and that in a deeper energy side, respectively. Note that the results derived for the trap distributions do not contradict our analysis where validity is assured for the r values ranging from 0.5 to unity. An alternative explanation for the inflection in the μ_p' curve might be possible by assuming a shallow-trap-type distribution plus a deep exponential tail. At present stage of investigation, both models could be raised with equal plausibility. The careful numerical calculation to simulate the μ_p' behavior must be needed for deriving what kind of trap distribution is actually present, although it is beyond focus of the present work.

Before concluding this section, we briefly address the validity of a simplification in expression for the SSPG parameter Γ from Eq. (23) to Eq. (24), upon which our analysis is developed for the determination of transport parameters. This can be easily tested by evaluating the second-order transport parameters. It is obvious from above discussion that $1/\tau_{np}'' = 1/\tau_p'' = 0$ and $\mu_n'' = 0$. The nonzero parameters are $1/\tau_n''$ and μ_p'', which are written by $\tau'/\tau_n'' = (1-\alpha)/2\alpha$ and $\mu_p''/\mu_n' = (T_v - T)/2Tbr$. Using the present results, the ratios τ'/τ_n'' and μ_p''/μ_n' are both amounted to be only less than several percent. This fact satisfactorily validates the application of the simplified expression (24).

IV. CONCLUSIONS

We have investigated theoretically deficiencies in the original low-field SSPG technique arising from the incorrect application of the perturbation expansion theory as well as the uncertain assumption of local charge neutrality. We have also developed an extended technique for accurate diffusion length measurement by removing these defects, and have presented experimental data for a-Si:H obtained as a function of the illumination level. We can summarize our results as follows.

(i) Applying the second-order perturbation theory precisely, it is shown that the SSPG parameter Γ, defined in the formulation for the small-signal current ratio measured in the experiment, is comprised of two components associated with the drift-current and the diffusion-current. The lack of the diffusion-current component is a defect in the previous SSPG theory.

(ii) The correction of the SSPG theory is highly needed in the relaxation-time regime where the space-charge effect becomes significant, while there is no difference between the results derived from the present and previous theories in the lifetime regime where ambipolar transport takes place.

(iii) When the lifetime regime is not met practically in the experiment, a severe overestimate of diffusion length will result from the SSPG data analysis assuming the local charge neutrality. This defect is inherent in the diffusion length measurement relying only on the SSPG technique.

(iv) Combining the SSPC and FRPC measurements, which determine the small-signal mobility-lifetime product and the small-signal lifetime, respectively, enables us to avoid such an experimental failure, and to extend the diffusion length measurement beyond the ambipolarity restriction.

(v) Experimental application of this technique to a-Si:H has revealed the correct value of diffusion length and its genuine light-intensity dependence in the intensity range $1-100$ mW/cm^2, which amply demonstrate serious errors in the previous evaluations.

(vi) The present combined technique has a merit of being able to determine the small-signal drift mobilities of both types of carriers. The results measured for a-Si:H with varying the illumination level can be well interpreted by assuming that transports of photogenerated electrons and holes are controlled by the trap states distributed in the gap, with a sharp and an exponential drop, respectively.

APPENDIX

Using the current continuity and periodicity, the SSPG current J, the sum of drift current $\sigma(x)E(x)$ and diffusion current $J_d(x)$, can be formulated in the following form:

$$J = \sigma_{\mathrm{av}}(E_0 + E^*) \ , \quad (A1)$$

where $\sigma_{\mathrm{av}} = 1/\langle 1/\sigma(x)\rangle$ denotes the average conductivity in the direction perpendicular to the grating fringes, and $E^* = \langle J_d(x)/\sigma(x)\rangle$ the field correction due to the local diffusion current $J_d(x)$. Ritter, Zeldov, and Weiser have analyzed the SSPG current on the groundwork of simple Ohm's law, that is, $J = \sigma_{\mathrm{av}} E_0$.[1,2] Evidently, such a treatment can be applied validly only when $J_d(x)$ is negligibly small. It is, however, evident that $J_d(x)$ should not be ignored in the context of the SSPG experiment that measures photocurrent generated by an optical illumination grating.

The average conductivity σ_{av} can be expressed in the

form of perturbation expansion. One has the expression up to the second-order correction

$$\sigma_{\rm av}=\sigma_0+\langle\sigma_1\rangle+(\langle\sigma_1\rangle^2-\langle\sigma_1^2\rangle)/\sigma_0 , \tag{A2}$$

when neglecting the minor correction term $\langle\sigma_2\rangle$. Similarly, the field correction E^* is given by

$$E^*=-\langle J_{d1}\sigma_1\rangle/\sigma_0^2 , \tag{A3}$$

where $J_{d1}=q(D_n' dN_1/dx-D_p' dP_1/dx)$. From Eqs. (A1)–(A3), we obtain the expression for β, the current ratio measured in the SSPG experiment

$$\beta=1+(\langle\sigma_1\rangle^2-\langle\sigma_1^2\rangle)/\sigma_0\langle\sigma_1\rangle-\langle J_{d1}\sigma_1\rangle/\sigma_0\langle\sigma_1\rangle E_0 . \tag{A4}$$

After some mathematical computations, the second and third terms on the right-hand side of Eq. (A4) are found to be represented by

$$(\langle\sigma_1\rangle^2-\langle\sigma_1^2\rangle)/\sigma_0\langle\sigma_1\rangle=-2\gamma_0^2\alpha\Gamma_e , \tag{A5}$$

$$\langle J_{d1}\sigma_1\rangle/\sigma_0\langle\sigma_1\rangle E_0=-2\gamma_0^2\alpha\Gamma_d , \tag{A6}$$

with the drift current component Γ_e and the diffusion-current component Γ_d given by Eq. (26).

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