

Energy-level dynamics of deep gap states in hydrogenated amorphous silicon under illumination

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Relaxation dynamics of deep defects, recently observed in modulated photocurrent and electron drift measurements, are explained in terms of random temporal fluctuations of localized-state energies. The fluctuations can be caused by a randomly changing microscopic configuration of excess charge carriers or by carrier annihilation processes. A model is developed which leads to dependences of the thermal emission energy upon temperature and optical bias, in good agreement with existing experimental data. [S0163-1829(96)06547-2]

I. INTRODUCTION

Apparently changing energy positions of deep gap states under the influence of optical excitation constitutes one aspect of the set of experimental observations on hydrogenated amorphous silicon (*a*-Si:H) that has become known by the name *defect relaxation*. Illumination reduces the depth of the state. Examples of such behavior have been presented by Zhong and Cohen¹⁻³ and by Han *et al.*,⁴ and a possible mechanism was discussed by Branz and Schiff.⁵ The phenomena here are being considered independently of the ‘‘other’’ cases of defect relaxation which involve a *deepening* of the trap. Characteristic features of the observed upward energy shifts are a linear temperature dependence and a logarithmic variation with the optical generation rate.¹

The above phenomena are strongly reminiscent of the apparent temperature dependence of the *a*-Si:H gap-level density of states which has been repeatedly reported in recent years,^{1,6-10} and which can be explained by making use of the slow random fluctuations of gap-state energies that are bound to occur in all disordered systems. Such fluctuations, which conform to the typical $1/f$ noise characteristics, do produce a distribution of thermal activation energies (DAE) for carrier release out of deep states which deviates from the distribution of equilibrium energies of those states. This difference not only explains the apparent temperature dependence, referred to above, but also has consequences for other material properties such as the equilibrium mobility¹¹ or the dispersive transport.¹² In the present paper, we will show that these fluctuations are capable of explaining the above-mentioned *defect relaxation* effects, and that it is very reasonable to assume that the optical excitation will in fact generate the type of energy fluctuations that are required.

The experimentally observed dependence of DAE's upon the light-beam intensity, i.e., upon the carrier generation rate,¹⁻⁴ implies a collective, many-electron nature of the effect. In an illuminated photosensitive medium, any carrier is surrounded by nonequilibrium electrons and holes, forming a Coulomb potential landscape which is random in both space and time. The spatial part has been used in various attempts in the literature¹³⁻¹⁷ to relate energy distributions of band-tail states to static random distributions of charged defects in amorphous semiconductors. However, by changing their positions, charge carriers will also cause temporal fluctuations of localized state energies. Since in amorphous materials

practically all carriers are localized, any change in position requires that a carrier first be released from a localized state. Therefore a characteristic frequency of these ‘‘Coulomb’’ fluctuations can be estimated from the frequency of release events in a sufficiently close neighborhood of a given state. Another possible origin of energy fluctuations is the process of carrier recombination. In this case both nonequilibrium phonons emitted during the electron-hole annihilation and structural changes caused by the latter process¹⁸ can be responsible for the fluctuations. Both Coulomb and recombination processes have been identified as sources of $1/f$ noise in disordered semiconductors.^{18,19} Consequently, we will propose a $1/f$ -type frequency dependence for the fluctuations.

II. DISTRIBUTION OF EFFECTIVE ACTIVATION ENERGIES

To calculate the effect of fluctuations on the thermal release of trapped carriers, we will use simple oscillatory motion as a model. The energy of a carrier, captured by an oscillating localized state, then changes with time as

$$E(t) = E_t + \Delta \cos(2\pi ft + \varphi_0), \quad (1)$$

where E_t is the average energy of the state, and Δ , f , and φ_0 are the amplitude, frequency, and initial phase of the oscillation. At any given time an ensemble of oscillating states with random values of the initial phase can be characterized by the following energetic distribution:

$$\gamma(E) = 2f \frac{dt(E)}{dE}, \quad E_t - \Delta < E < E_t + \Delta. \quad (2)$$

Substituting Eq. (1) into Eq. (2) yields,

$$\gamma(E) = \frac{1}{\pi\Delta \left[1 - \left(\frac{E - E_t}{\Delta} \right)^2 \right]^{1/2}}, \quad E_t - \Delta < E < E_t + \Delta. \quad (3)$$

The oscillations of localized-state energies thus make a single level spread inhomogeneously within the interval from $E_t - \Delta$ to $E_t + \Delta$. The question can then be asked: what will be the average activation energy for carrier release from such oscillating localized state? The probability of a thermally activated carrier jump is determined by both the effective

energy distribution of the state and the probability to get sufficient energy from a phonon. The latter is determined by the Boltzmann factor, and the average activation energy E_a for the carrier jump from an oscillating state can therefore be evaluated as

$$\exp\left(-\frac{E_a}{kT}\right) = \frac{1}{\pi\Delta} \int_{E_t-\Delta}^{E_t+\Delta} dE \frac{\exp\left(-\frac{E}{kT}\right)}{\left[1 - \left(\frac{E-E_t}{\Delta}\right)^2\right]^{1/2}}. \quad (4)$$

The integrand in Eq. (4) reveals a sharp maximum close to the lower bound, yielding

$$E_a \cong E_t - \Delta. \quad (5)$$

Thus, oscillations of the localized-state energies with amplitude Δ , are equivalent to an upward shift of the level energies by the same value Δ , as far as thermal release of carriers is concerned.

It should be reasonable to assume that the amplitude of the random temporal fluctuations in disordered materials would be related to the time scale within which the fluctuations occur, and hence show an inverse-frequency behavior. Considered over longer times, any characteristic of a random system is subjected to stronger fluctuations. More specifically, it was shown²⁰ that thermal-equilibrium diffusion and/or recombination of charged particles leads to a noise spectrum which can be described by the inverse square-root dependence upon the characteristic frequency, $\Delta \propto f^{-1/2}$, while the dispersive transport and recombination yield the well-known $1/f$ noise.^{19,20} Therefore the function

$$\Delta(f) = \Delta_0 \left(\frac{f_0}{f}\right)^\alpha, \quad 0.5 \leq \alpha \leq 1.0, \quad (6)$$

where Δ_0 and f_0 characterize the amplitude and the time scale of the fluctuations, should be a reasonable ansatz for studying the effect of the fluctuations. The values of both Δ_0 and f_0 can be very different in the presence and absence of external excitation. First we consider the effect of fluctuations on small-signal carrier transport when a sample remains in equilibrium. Under these conditions the characteristic frequency f_0 represents a material parameter and in line with experimental observations,²¹ it is anticipated that f_0 has a value on the order of 1 Hz for a -Si:H. The prefactor Δ_0 must account for the temperature dependence of fluctuations. The choice of a particular dependence is, of course, somewhat arbitrary. However, it is demonstrated below that this choice does not strongly affect the final results as long as the temperature dependence of Δ_0 is weaker than the exponential. On this basis we will use a linear dependence $\Delta_0 = kT$.

A carrier undergoes energy fluctuations together with a localized state until it is released after the time t_r ,

$$t_r = \frac{1}{\nu_0} \exp\left(\frac{E_a}{kT}\right). \quad (7)$$

Therefore, the lowest fluctuation frequency f (and the largest amplitude) that will contribute will correspond to the inverse release time $f = 1/t_r$. Substituting Eq. (7) into Eq. (6) and

using Eq. (5) yields the following relation between the average energy of a localized state E and its effective activation energy E_a :

$$E = E_a + kT \left(\frac{f_0}{\nu_0}\right)^\alpha \exp\left(\frac{\alpha E_a}{kT}\right). \quad (8)$$

The effective activation energy is, in general, very sensitive to the temperature, but with the crucial term $\exp(\alpha E_a/kT)$ being completely independent, our choice of temperature dependence for the prefactor Δ_0 .

The distribution of effective activation energies $g_{\text{eff}}(E_a)$ is related to the distribution of average energies $g(E)$, i.e., to the standard density-of-states function, by the following expression:

$$\begin{aligned} g_{\text{eff}}(E_a) &= g[E(E_a)] \frac{dE(E_a)}{dE_a} \\ &= g[E(E_a)] \left[1 + \alpha \left(\frac{f_0}{\nu_0}\right)^\alpha \exp\left(\frac{\alpha E_a}{kT}\right) \right]. \end{aligned} \quad (9)$$

Since the characteristic frequency of the fluctuations, f_0 , is much less than the attempt-to-escape frequency, ν_0 , the second term in the right-hand side of Eq. (8) is much less than the first one at high temperatures. Concomitantly, at high temperatures the effective activation energy E_a is very close to the average energy E , and the distribution $g_{\text{eff}}(E_a)$ just follows the function $g(E)$. At lower temperatures the values E_a and E are similar only for sufficiently shallow states. For states with higher average energies, E_a is much smaller than E . As a result, the distribution $g_{\text{eff}}(E_a)$ coincides with $g(E)$ only down to the energy $E_b(T) \cong kT \ln(\nu_0/f_0)$. Effective activation energies of states with average energies $E > E_b$ are compressed into a relatively narrow band around the energy E_b . Temperature dependences of the effective density of states are illustrated on Fig. 1 for (a) the equilibrium $\alpha=0.5$ case, and (b) the dispersive $\alpha=1$ mode of the fluctuations. Comparing the two sets of curves shows that the function $g_{\text{eff}}(E_a)$ is not very sensitive to the value of α , though the temperature-dependent edge of the distribution becomes sharper with increasing α . Therefore, in the following sections we consider enhanced fluctuations only for the genuine $1/f$ noise with $\alpha=1$.

III. PHOTOINDUCED SHIFT OF THE EFFECTIVE ACTIVATION ENERGY

The random fluctuations discussed in Sec. II can be generated by photoinduced changes in electron and hole densities, either through Coulomb interactions between moving charges, or when the carriers recombine. First we consider the Coulomb mechanism of fluctuations. Estimating the parameter f_0 as the frequency ν of carrier release events within a volume V , surrounding the localized state, and the parameter Δ_0 as the characteristic Coulomb energy $E_C = \kappa e^2/\epsilon V^{1/3}$ of the electron-electron interaction within this volume yields

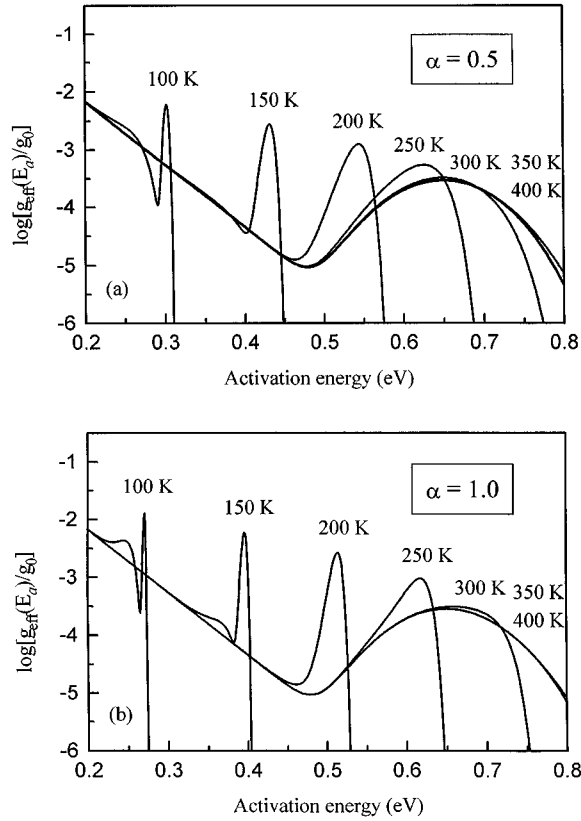


FIG. 1. Temperature-dependent distributions of the localized-state activation energies for (a) the equilibrium $\alpha=0.5$ and (b) dispersive $\alpha=1.0$ diffusion-controlled noise spectra. $g(E) = g_0 \{ 0.999 \exp(-E/E_0) + 0.001 \sqrt{\pi} (E_0/E_1) \exp[-(E-E_1)^2/E_1^2] \}$, where $E_0=0.04$ eV, $E_1=0.65$ eV, $E_1=0.08$ eV, and $\nu_0/f_0=10^{12}$.

$$\begin{aligned} \Delta_0 f_0 &= E_C N V \nu = \frac{\kappa e^2}{\varepsilon V^{1/3}} N V \nu_0 \exp\left(-\frac{E_a}{kT}\right) \\ &= \frac{\kappa e^2 N V^{2/3}}{\varepsilon} \nu_0 \exp\left(-\frac{E_a}{kT}\right), \end{aligned} \quad (10)$$

where N is the density of carriers, T the temperature, ν_0 the attempt-to-escape frequency, e the elementary charge, ε the dielectric permittivity, κ the electric constant, and k the Boltzmann constant. Substituting Eqs. (10) and (7) with $t_r^{-1}=f$ into Eq. (6) for $\alpha=1$ yields

$$\Delta = \frac{\kappa e^2 V^{2/3} N}{\varepsilon}. \quad (11)$$

Thus the Coulomb mechanism of fluctuations leads to a linear dependence of the energy shift Δ upon the carrier density. The use of Eq. (7) does not imply that one should not also consider the bandtail states (of which there are a large number in the volume) as the source of the fluctuations. The characteristic frequency of these fluctuations remains, nevertheless, determined by the carrier release time from the deep states, since this is the slowest process involved. In equilibrium practically all carriers are localized in deep traps and, to reach a bandtail state, a carrier must first be released from a deep trap. Of course, taking the bandtail states into account

would somewhat change the kinetics of the carrier densities at short times before the equilibrium is established, but the overall time to reach equilibrium would remain essentially unchanged. Therefore, the lowest fluctuation frequency f , which will have the largest amplitude, is correctly evaluated as the inverse release time $f=1/t_r$.

For the recombination mechanism of fluctuations the values Δ_0 and f_0 should be estimated as the energy E_R released in a recombination event and the rate of recombination within the volume V , respectively. The product $\Delta_0 f_0$ must also account for the probability γ that a recombination event will trigger a structural rearrangement,²²

$$\Delta_0 f_0 = \gamma E_R V R_0 N_c N = \gamma E_R V R_0 N^2 \exp\left(-\frac{E_a}{kT}\right), \quad (12)$$

where R_0 is the constant of recombination of free electrons with holes, and N_c the density of free electrons. Equation (12) is written assuming that (i) the density of free carriers is much less than the total carrier density and these two densities are related as $N_c = N \exp(-E_a/kT)$ and (ii) the total densities of electrons and holes are equal. Using Eqs. (7) and (12) in Eq. (6) now leads to an oscillation amplitude and hence energy shift Δ , which is proportional to the squared total density of carriers,

$$\Delta = \frac{\gamma E_R V R_0 N^2}{\nu_0}. \quad (13)$$

To estimate the time evolution of the photoinduced energy shifts, one has to evaluate the time dependence of N . The kinetics of the carrier density is governed by the carrier generation G and recombination rates,

$$\begin{aligned} \frac{dN(t)}{dt} &= G(t) - R_0 N_c(t) N(t) = G(t) \\ &\quad - R_0 \exp\left(-\frac{E_a}{kT}\right) [N(t)]^2. \end{aligned} \quad (14)$$

Substituting Eq. (5) and either Eq. (11) or Eq. (13) into Eq. (14) yields equations describing the dynamics of the carrier density for the Coulomb,

$$\frac{dN(t)}{dt} = G(t) - R_0 [N(t)]^2 \exp\left[-\frac{E_t - \chi' N(t)}{kT}\right], \quad (15)$$

and for the recombination mechanism of fluctuations,

$$\frac{dN(t)}{dt} = G(t) - R_0 [N(t)]^2 \exp\left\{-\frac{E_t - \xi' [N(t)]^2}{kT}\right\}, \quad (16)$$

where

$$\chi' = \frac{\kappa e^2 V^{2/3}}{\varepsilon}, \quad \xi' = \frac{\gamma E_R V R_0}{\nu_0}. \quad (17)$$

In the following we consider a step-function generation rate,

$$G(t) = G_0, \quad 0 \leq t \leq t_0, \quad G(t) = 0, \quad t < 0, \quad t > t_0. \quad (18)$$

It is convenient to rewrite Eqs. (15) and (16) using the following dimensionless variables:

$$\tau = t\sqrt{G_0 R}, \quad n = N \left(\frac{R}{G_0} \right)^{1/2}, \quad \chi = \frac{\chi'}{kT} \left(\frac{G_0}{R} \right)^{1/2},$$

$$\xi = \frac{\xi'}{kT} \frac{G_0}{R}, \quad (19)$$

where $R = R_0 \exp(-E_i/kT)$. Substituting Eq. (19) into Eqs. (15) and (16) yields

$$\frac{dn(\tau)}{d\tau} = 1 - [n(\tau)]^2 \exp[\chi n(\tau)], \quad (20)$$

for the Coulomb noise, and

$$\frac{dn(\tau)}{d\tau} = 1 - [n(\tau)]^2 \exp\{\xi [n(\tau)]^2\} \quad (21)$$

for the recombination noise. The effective activation energy E_a for these two kinds of noise should then be written as

$$E_a = E_i - kT\chi n \quad (22)$$

and

$$E_a = E_i - kT\xi n^2, \quad (23)$$

respectively. Equations (20)–(23) show that energetic fluctuations can affect kinetics of both the carrier density and the activation energy only if the parameters χ and ξ are not much less than unity. Assuming values of the parameters $V = 10^{-17} \text{ cm}^3$, $E_i = 0.65 \text{ eV}$, $E_R = 1 \text{ eV}$, $\gamma = 10^{-3}$, $\varepsilon = 12$, $\nu_0 = 10^{11} \text{ s}^{-1}$, $R_0 = 10^{-7} \text{ cm}^3 \text{ s}^{-1}$, and $G_0 = 10^{15} \text{ cm}^{-3} \text{ s}^{-1}$, one obtains $\chi \approx 10^{-3}$ and $\xi \approx 3 \times 10^{-3}$ for $T = 300 \text{ K}$ and $\chi \approx 1$ and $\xi \approx 10^3$ for $T = 200 \text{ K}$. Both parameters increase with decreasing temperature and/or with increasing generation rate. At first glance the recombination mechanism of fluctuations makes the dominant contribution to the $1/f$ noise. However, this conclusion would have to be supported by a more detailed analysis of the problem before it can be relied upon. Therefore we will continue to consider both mechanisms.

IV. RESULTS AND DISCUSSION

First we consider steady-state solutions of Eqs. (20) and (21). At small values of the fluctuation parameters χ and ξ ($\chi \ll 1$, $\xi \ll 1$) both equations yield $n_{\text{st}} = 1$. For $\chi \gg 1$ and $\xi \gg 1$ approximate solutions of these equations are $n_{\text{st}} = (2/\chi) \ln \chi$ and $n_{\text{st}} = \sqrt{\ln \xi/\xi}$. The steady-state carrier density N_{st} is therefore

$$N_{\text{st}} = \left(\frac{G_0}{R} \right)^{1/2}, \quad \chi \ll 1; \quad N_{\text{st}} = \frac{2kT}{\chi'} \ln \left[\frac{\chi'}{kT} \left(\frac{G_0}{R} \right)^{1/2} \right], \quad \chi \gg 1 \quad (24)$$

for the Coulomb fluctuations, and

$$N_{\text{st}} = \left(\frac{G_0}{R} \right)^{1/2}, \quad \xi \ll 1; \quad N_{\text{st}} = \left[\frac{kT}{\xi'} \ln \left(\frac{\xi'}{kT} \frac{G_0}{R} \right) \right]^{1/2}, \quad \xi \gg 1 \quad (25)$$

for the recombination-induced fluctuations. In both instances there is a transition from the ‘‘normal’’ square-root depen-

dence of the carrier density upon the generation rate to a much weaker logarithmic one and a simultaneous weakening of the temperature dependence of N . Substituting Eqs. (24) and (25) into Eqs. (11) and (13) gives steady-state values for the activation energies,

$$E_a^{(\text{st})} = E_i, \quad \chi \ll 1; \quad E_a^{(\text{st})} = kT \ln \left[\left(\frac{kT}{\chi'} \right)^2 \frac{R_0}{G_0} \right], \quad \chi \gg 1, \quad (26)$$

$$E_a^{(\text{st})} = E_i, \quad \xi \ll 1; \quad E_a^{(\text{st})} = kT \ln \left(\frac{kT}{\xi'} \frac{R_0}{G_0} \right), \quad \xi \gg 1. \quad (27)$$

Equations (26) and (27) show that at low temperatures or/and at high generation rates, i.e., when χ and ξ are large, the steady-state effective activation energy is not related to its value in the absence of fluctuations. One should also emphasize the practically linear temperature dependence of $E_a^{(\text{st})}$ and its logarithmic dependence upon the generation rate, both in good agreement with existing experimental data.^{1–3} We also present formulas for the free-carrier density which determines the temperature and generation-rate dependences of the steady-state photocurrent. One obtains

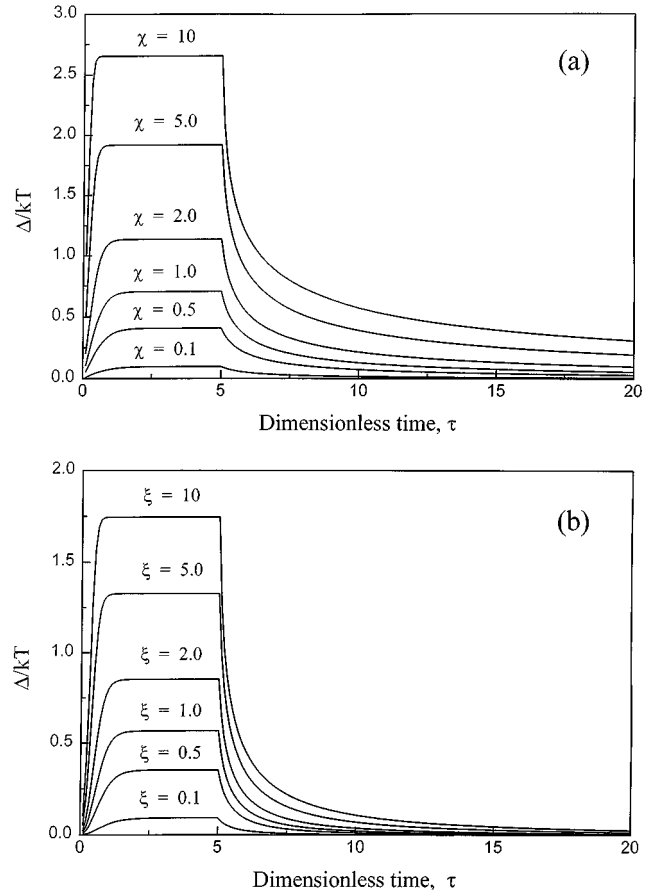


FIG. 2. Time dependence of the effective activation energy of deep localized states after application and cessation of the bias light for (a) Coulomb and (b) recombination mechanisms of fluctuations.

$$N_c^{(st)} = \left(\frac{G_0}{R_0}\right)^{1/2} \exp\left(-\frac{E_t}{2kT}\right), \quad \chi \ll 1;$$

$$N_c^{(st)} = \frac{\chi'}{2kT} \frac{G_0}{R_0} \left\{ \ln \left[\frac{\chi'}{kT} \exp\left(\frac{E_t}{kT}\right) \left(\frac{G_0}{R_0}\right)^{1/2} \right] \right\}^{-1}, \quad \chi \gg 1$$
(28)

for Coulomb fluctuations, and

$$N_c^{(st)} = \left(\frac{G_0}{R_0}\right)^{1/2} \exp\left(-\frac{E_t}{2kT}\right), \quad \xi \ll 1;$$

$$N_c^{(st)} = \left(\frac{\xi'}{kT}\right)^{1/2} \frac{G_0}{R_0} \left\{ \ln \left[\frac{\chi'}{kT} \exp\left(\frac{E_t}{kT}\right) \frac{G_0}{R_0} \right] \right\}^{-1/2}, \quad \xi \gg 1$$
(29)

for recombination-induced fluctuations. When fluctuations become important, a square-root dependence of the photocurrent upon the generation rate is changed to an almost linear dependence, simulating a transition from bimolecular to monomolecular recombination.

Kinetics of the effective activation energy upon application of the step-function generation of Eq. (18) is illustrated in Figs. 2(a) and 2(b) for both mechanisms of fluctuations and for various values of the fluctuation parameters. An obvious asymmetry is seen between characteristic relaxation

times after applying and removing the bias light, with the latter being much longer than the former, especially for Coulomb fluctuations. This agrees with the experimental observation of Cohen and Zhong.³ Also noteworthy is that our estimated shifts of several times kT are of the order of the experimentally observed shifts^{3,4} of about 0.1 eV.

V. CONCLUDING REMARKS

In this contribution we have shown that the low-frequency random fluctuations can account for the observation of optically induced shifts of deep-state energies. However, it should be pointed out that the model has only been developed for monoenergetic states, and that both a distribution of defect states and the influence of band tails will to some extent modify the above functional dependences. These considerations, as well as the application of the fluctuations concept to standard models of photoconductivity controlled by distributions of localized states, will have to be addressed in subsequent work.

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