# ELECTRICAL CONDUCTIVITY INDUCED IN INSULATORS BY PULSED RADIATION[*]

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## ABSTRACT

The minimum prompt photoconductivity induced by pulses of x rays, gamma rays, and energetic electrons in various amorphous and disordered insulating organic and inorganic materials is predicted on the basis of data for the scattering of hot electrons in solids and the band gap for insulators. For total doses of $3 \times 10^4$ to $30 \times 10^4$ rad or greater, the minimum prompt photoconductivity is predicted to be linear with dose rate, $\dot{\gamma}$, and is given by

$$\sigma(\Omega^{-1}\text{cm}^{-1}) = 5 \times 10^{-19} \, \rho_0 \dot{\gamma}/E_g^2,$$

where $\rho_0$ is the density $(\text{g/cm}^3)$ and $E_g$ is the optical band gap (eV). This formula agrees well with data for a variety of plastics, mica, and borosilicate glass under widely different irradiation conditions. The formula considerably underestimates absolute values of prompt conductivities observed for $Al_2O_3$, $MgO$, and certain plastics, because the model does not hold for ordered materials.

## I. INTRODUCTION

Insulating materials subjected to high-intensity radiation bursts exhibit a measurable, induced, transient conductivity.[1-5] In general, calculating the conductivity induced by intense irradiation is very complicated. Here, we derive a rather simple formula for calculating the minimum prompt conductivity of insulators induced by x rays (or electrons) at dose rates greater than about $10^{17}$ photons $\text{cm}^{-3} \text{ s}^{-1}$ and at pulse times in the nanosecond to microsecond range. The whole argument is based on the simplicity of the physics of the problem in the first few nanoseconds or, sometimes, even the first few microseconds. At these early times, only hot electrons contribute to the conductivity because low-energy electrons are quickly trapped ($\sim 10^{-10}$ s)[5] and generally have negligible mobility.[6] The inverse process of emptying shallow traps can be completely ignored because it is a relatively slow process with a time constant of approximately $10^{-5}$ s, which causes delayed photoconductivity.[5] We do not expect this model to apply to highly ordered crystalline materials. It can, at best, apply only to glassy or disordered materials for which the conductivity of low-energy electrons can be ignored. Clearly, under steady-state irradiation, the density of low-energy electrons is expected (and is observed) to increase with time and eventually to make a significant or even dominant contribution to the conductivity. The question is,

how long can thermalized electrons be ignored? For borosilicate glass, this time appears to be of the order of $10^{-6}$ s. For some materials, it is probably much too short (say, $<10^{-12}$ s) to allow measurement of the conductivity of hot electrons, which precludes comparison with our model.

We make calculations specifically for glassy and plastic materials, which have band gaps of about 5 eV, and then comment on the general application to other materials.

## II. THEORY

Each absorbed photon creates an excited electron and a hole. Because holes in insulators are quickly trapped and are generally relatively immobile, we ignore them. Following the initial excitation is a cascade process in which plasmons and secondary electrons are produced. Thus, in the steady state, there are electrons in the conduction bands ranging in energy from $(h\nu - E_g)$, where $h\nu$ is the photon energy and $E_g$ is the band gap energy, down to the conduction band minimum. Most of the energy of the high-energy electrons is probably initially dissipated in plasmon excitation; however, the plasmons decay into electron-hole pairs so that the final result is essentially the same.[7] An error factor of less than two in the steady-state density of hot electrons results from ignoring plasmon excitation. This is well within the accuracy of the simple model proposed here. Because one high-energy primary electron of energy $(h\nu - E_g)$ can produce two electrons, each with nearly half the primary electron energy, the number of excited electrons per unit energy in steady state increases with decreasing secondary electron energy. About half of the hot electrons lie within an energy range of the order of $E_g$ above the conduction band minimum. Figure 1 shows the electronic band structure for a disordered system and illustrates that a fairly wide energy range for electrons of negligible mobility can be expected in glassy materials.

Ignoring plasmon production, each absorbed photon of energy $h\nu$ leads to the excitation of approximately $h\nu/E_g$ conduction electrons because of electron-hole pair production. Thus, the steady-state rate of electron excitation to the conduction bands is $fh\nu/E_g$, where $f$ is the rate of photon absorption per unit volume. Then the steady-state electron concentration in the conduction bands is

$$N = fh\nu\tau/E_g , \qquad (1)$$

where $\tau$ is the lifetime of an electron. The conductivity is

$$\sigma = Ne^2\tau/m$$
$$= fh\nu e^2\tau^2/mE_g . \qquad (2)$$

All the quantities needed for Eq. (2) are known except $\tau$. A rough estimate of $\tau$ may be obtained using the uncertainty principle and experimental information on the uncertainty in the energy of hot

electrons, $\delta E$. Photoelectron energy-distribution measurements of electrons having energies in the range of 5 to 10 eV indicate an uncertainty in energy of the order of 0.2 eV.[9] Thus,

$$\tau \approx \frac{h}{\delta E} \approx 3 \times 10^{-15} \text{ s.} \qquad (3)$$

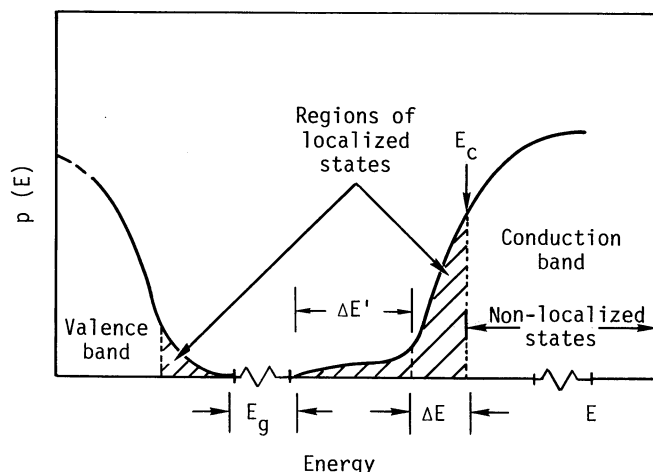This value of $\tau$ leads to a close estimate of the minimum prompt photoconductivity.



Fig. 1. Hypothetical electron density of states vs energy for a glassy insulator (after Mott and Davis[8]). The density of states in the conduction band is that suggested when the states are p-like or d-like, as in glass. The energy $E_c$ corresponds to the mobility shoulder, below which the mobility is expected to drop by $10^3$ or more. The region of width $\Delta E$ ($\sim 0.2$ eV) is a region of localized states. The additional band tailing of width $\Delta E'$ arises from fluctuations in density. The $E_g$ region is not to scale but is typically 2 to 6 eV.

Another estimate of $\tau$ is available from an analysis of the scattering of hot electrons as observed in photoemission experiments. Those experiments show that the mfp for electron scattering is typically about 15 Å for electrons with energies 1 to 3 $E_g$ above the conduction-band minimum in semiconductors and insulators and also for electrons 5 to 10 eV above the Fermi energy in metals.[10,11] Assuming that such energetic electrons can be treated as free electrons, the lifetime can be estimated from

$$\tau = \frac{\ell}{v}, \qquad (4)$$

where $v$ is the velocity and $\ell$ is the mfp. The lifetime for conduction-band electrons estimated from Eq. (4) is $10^{-15}$ s for energies of 6 eV. Because the mfp for electrons at energies more than a band gap above the conduction-band minimum is determined almost entirely by electron-electron scattering, the lifetime calculated from Eq. (4) is that for energy loss by pair production. Thus, those electrons lying within a few $E_g$ above the conduction-band minimum will, in a time of the order of $10^{-15}$ s, undergo one last process of pair production in which they lose energy of the order of $E_g$. Because only about half of the excited electrons lie within a few $E_g$ above the conduction band minimum (disregarding nearly thermalized electrons, which we discuss later), this

is equivalent to a mean lifetime, $\tau$, in the conduction bands of approximately $2 \times 10^{-15}$ s.

We can rewrite Eq. (4) by expressing v in terms of E and m, the conduction electron energy and mass, respectively. Thus

$$\tau = 2\ell(m/2E)^{1/2}, \qquad (5)$$

where an additional factor of two has been included to account for the approximately equal number of higher-energy electrons that have more than one pair-production process left to undergo. As indicated previously, the energy of an electron undergoing its last electron-electron scattering event has an energy of order $E_g$. Taking $E = E_g$ in Eq. (5) and then substituting for $\tau$ in Eq. (2) yields

$$\sigma \cong 200 \ \dot{\gamma}\rho_0 \ (e\ell/E_g)^2, \qquad (6)$$

where $\dot{\gamma}$, the radiation dose rate in rads/s, is used instead of $f h\nu$, and $\rho_0$ is the mass density of the insulator (g/cm$^3$). Taking $\ell = 15 \times 10^{-8}$ cm and assuming $E_g$ is given in electron volts, the photoconductivity is

$$\sigma(\text{ohm}^{-1} \text{ cm}^{-1}) = 4.5 \times 10^{-19} \ \rho_0 \dot{\gamma}/E_g^2. \qquad (7)$$

Equation (7) correctly predicts a linear dependence of conductivity with dose rate. It assumes that low-energy electrons are quickly trapped or have negligible mobility, and it ignores the thermal excitation of electrons from trapping states. Thus, Eq. (7) provides a lower bound to the conductivity.

### III. COMPARISON WITH EXPERIMENTAL DATA

Approximate bounds on the conductivity expected from a variety of insulators may be obtained by assuming $E_g$ varies from 2 to 8 eV and $\rho_0$ varies from 1 to 4 g/cm$^3$. Substituting these values into Eq. (7) yields the theoretical range of conductivity shown in Fig. 2. The theoretical curves bracket the recent data of Lee et al.[2] for low-density polyethylene, several fluorocarbons, polystyrene, polyvinylidene chloride, polyvinylidene fluoride, polyvinyl chloride, and mica, all taken at dose rates of $10^9$ to $10^{11}$ rads/s. The theoretical curves in Fig. 2 also bracket the data of Weingart et al.[1] for epoxy, polyethylene terepthalate, and polymide as well as the pulsed reactor data of Harrison et al.[3] for polyisobutylene and polystyrene. In particular, for borosilicate glass irradiated with 10-keV x rays at dose rates of $5.9 \times 10^{10}$ rad/s ($1.25 \times 10^{21}$ photons cm$^{-3}$ s$^{-1}$), the calculated conductivity (using $E_g$ = 5 eV as the effective band gap for glass) is $2.8 \times 10^{-9}$ $\Omega^{-1}$cm$^{-1}$. This value is in good agreement with the experimental value[1] of $3.67 \times 10^{-9}$ $\Omega^{-1}$cm$^{-1}$.

Equation (7) underestimates the photoconductivity data of Pomerantz[13] for MgO of van Lint et al.[14] for Al$_2$O$_3$, and of Culler and Rexford[15] for quasistatic irradiation data for various glasses and for certain plastics at various dose rates.

Finally, we used Eq. (7) to estimate the prompt 13-MeV electron-induced conductivity of z-cut quartz and compared the result with the measured value of Okabe et al.,[4] as extrapolated to 5 μs (the experimental pulse width). Figure 3 shows this extrapolation and Fig. 2 shows the conductivity obtained.

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Fig. 2 (scientific plot)

**Legend:**
- A Low-density polyethylene
- B Tefzel*
- C Durasan
- D Kel-F, amorphous
- E Kel-F, crystalline
- F Polystyrene
- G Polypropylene
- H Polyvinylidene chloride
- I Polyvinyl chloride
- J Polyvinylidene fluoride
- K Mica

Y-axis: Photoconductivity — $\Omega^{-1}$ cm$^{-1}$ ($10^{-6}$, $10^{-8}$, $10^{-10}$, $10^{-12}$, $10^{-14}$, $10^{-16}$, $10^{-18}$)

X-axis: Dose rate — rad/s ($10^{-2}$, $10^0$, $10^2$, $10^4$, $10^6$, $10^8$, $10^{10}$, $10^{12}$, $10^{14}$, $10^{16}$)

Plot labels: MgO[12] 1.3 MeV-electrons; $Al_2O_3$, 33-MeV electrons[1]; 3.25 µs[14]; Polytetrafluoroethylene[1]; Polytetrafluoroethylene[12]; Polyimide[1]; Polyimide[12]; Polyethylene[1]; Borosilicate glass[1]; Epoxy[1]; Z-cut, quartz[4] 13 MeV-electrons; Polyethylene terepthalate[1]; Polyisobutylene[3]; Polystyrene[3]; 10-keV, x rays 40 ns; $SiO_2$,glass[15]; 150°C; 29°C; Glass, 1723[15]; $Co_{60}$-γ; Glass, 8871[15]; Theoretical bounds, from Eq. (7)
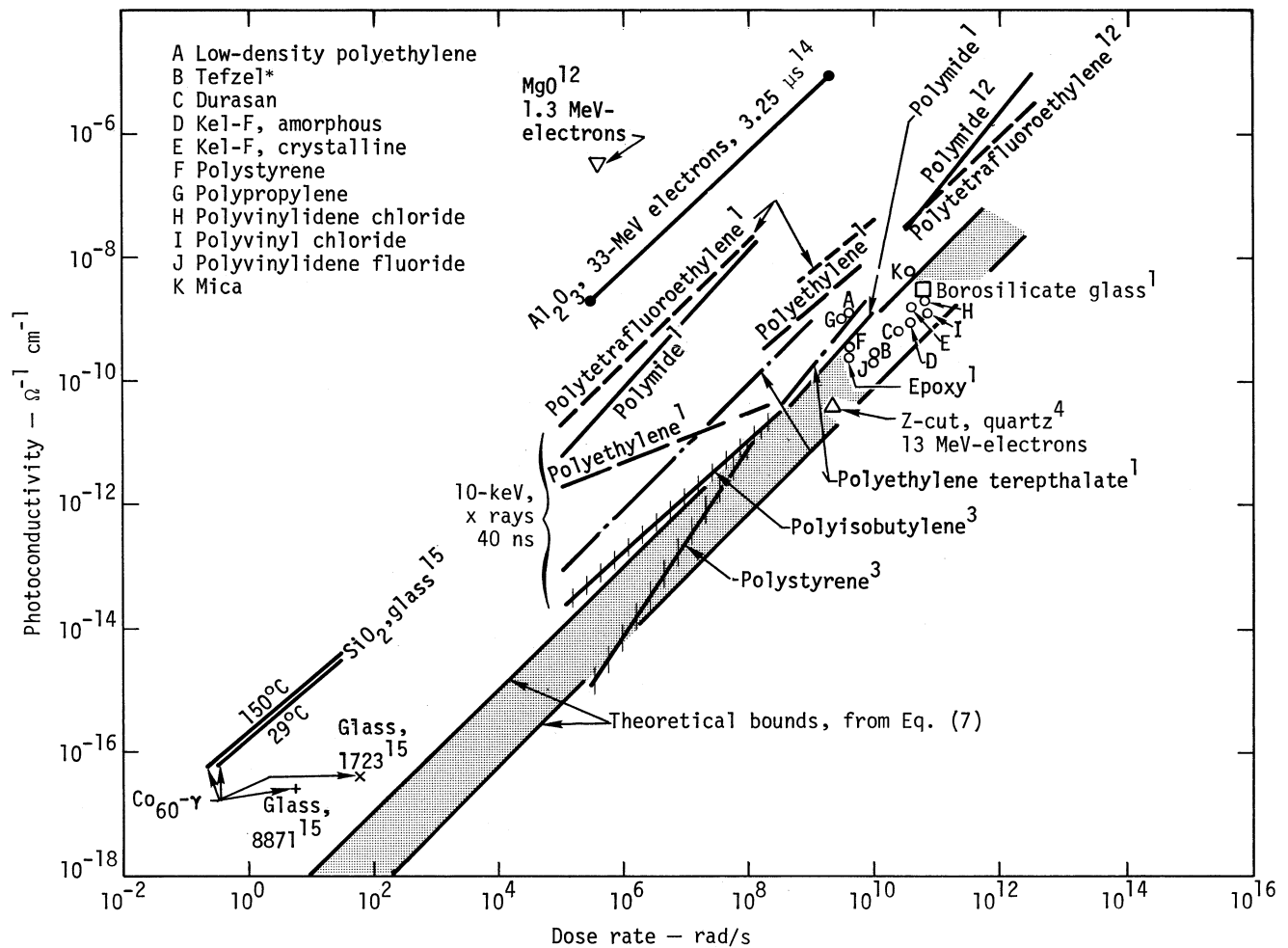
Fig. 2. Theoretical and experimental photoconductivities vs dose rates for various insulators. (*Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Energy Research and Development Administration to the exclusion of others that may be suitable.)

## IV. DISCUSSION

Our estimates of minimum photoconductivity use hot-electron scattering information inferred from photoemission experiments and estimates of hot-electron concentrations based on an energy $E_g$ for electron-hole pair production. Previous independent estimates either assumed an energy of 15 eV ($\sim$3 $E_g$) for electron-hole pair production[1] or separately inferred the mobility from phonon scattering theory.[13] The latter is inappropriate for hot electrons in glassy and plastic materials, and the proper threshold choice for pair production in disordered materials is closer to $E_g$ than 3 $E_g$.[16] For pure $SiO_2$, it has been shown that the threshold for pair production is 8.8 eV and that this energy is probably a more accurate value of the bandgap than the value of 9 eV determined from optical measurements.[17] Thus our choice of $E_g$ for both the band gap and the threshold for pair production in borosilicate glass is strongly supported by direct experimental measurements on related materials[17] as well as on chemically distinct but highly disordered materials.[16]

Considering the simplicity of the model, agreement was achieved with conductivity data for a number of glassy and amorphous materials subjected to a wide range of irradiation rates in different experiments. It must be partly fortuitous. Nonetheless, the close agreement of calculation and experiment presents strong support for the essential correctness of the model for amorphous materials at sufficiently short times after the onset of irradiation. Furthermore, the model suggests that all amorphous insulators having approximately the same band gap will exhibit the same induced conductivity under the same conditions of high-intensity pulsed irradiation. That is just what was found by Weingart et al.[1] It would not be expected if the conductivity were determined by impurity energy levels and concentrations and the various scattering mechanisms that typically determine the mobility and conductivity of thermalized electrons.

We must now consider some of the assumptions implicit in the model. The primary assumption is that only those electrons participating in the cascade process are treated as free electrons contributing to the conductivity. All electrons near the conduction-band minimum are assumed to be so quickly trapped or, more likely and more importantly, to have such a low mobility that their contribution to the conductivity is negligible. The agreement of the model with the experimental data provides some support of this conjecture.
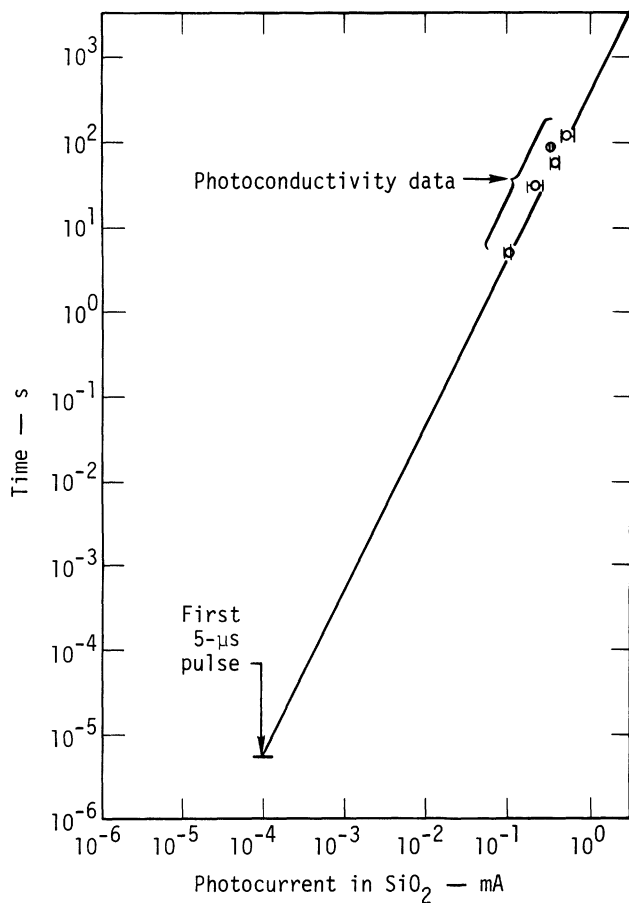
Time — s

$10^3$
$10^2$
$10^1$
$10^0$
$10^{-1}$
$10^{-2}$
$10^{-3}$
$10^{-4}$
$10^{-5}$
$10^{-6}$

Photoconductivity data →

First
5-μs
pulse

$10^{-6}$ $10^{-5}$ $10^{-4}$ $10^{-3}$ $10^{-2}$ $10^{-1}$ $10^0$

Photocurrent in $SiO_2$ — mA

Fig. 3.  Total irradiation time vs photo current induced by 13-MeV electrons impinging upon z-cut quartz (data from Okabe et al.[4]). The photoconductivity data is extrapolated to the first 5-μsec pulse.

A calculation of the mobility $\mu$ of the hot electrons yields

$$\mu = \frac{e\tau}{m} = 3.5\, cm^2 V^{-1} s^{-1}. \qquad (8)$$

The mobility estimated with the present model is about as low as it can be and still retain the concept of mobility.[6] It is, nonetheless, much greater than the mobility for thermalized electrons in the region of band tailing. In the band-tailing region, which consists of quasi-localized levels below the mobility gap, carrier motion must be described as phonon-assisted hopping. In this region, the mobility drops sharply to $10^{-2} cm^2 V^{-1} s^{-1}$ or lower.[6,18] There are no measurements or even reliable estimates of the electron mobility in glasses and amorphous insulators in the energy range of a few electron volts above the conduction-band minimum. However, based on the extremely low mobilities measured for thermalized electrons in some insulating materials, it appears likely that the mobility in glassy insulators is usually very low, even for electrons within a few electron volts of the conduction-band minimum. This is not always the case. Materials with negligible band tailing, mainly those that are very pure, even though amorphous, may have appreciable mobilities. Thermalized electrons have been found to have relaxation lifetimes of $10^{-8}$ s and drift mobilities of 20 $cm^2 V^{-1} s^{-1}$ in pure amorphous $SiO_2$.[18] However, as noted by Hughes,[18] trapped holes cause significant shortening of the lifetime when the accumulated dose

to the sample exceeds about 20 rads. Because the total dose considered here is at least two orders of magnitude greater than 20 rads, and the materials studied were neither ultrapure nor highly ordered, the more typical low mobilities for thermalized electrons in disordered amorphous materials seems more likely.

We used the same τ for both the lifetime of a hot electron and the relaxation time. It takes several τ for the drift velocity to have its usual meaning. Here, there is no steady-state drift; a hot electron is accelerated by an electric field for its lifetime, collides, and "disappears." Nonetheless, the collection of hot electrons exhibits a conductivity. Because the greatest contribution to this conductivity comes from the large number of hot electrons at the end of the cascade process, the lifetime of these electrons gives a reasonable measure of the conductivity even though (the fewer) higher-energy hot electrons might have much shorter lifetimes. The use of this hot-electron lifetime for a relaxation lifetime where no time-drift velocity exists produces at most an error of a factor of two in the calculated conductivity.

The mobility, as calculated in Eq. (8) using the hot-electron lifetime, is perhaps best thought of as an equivalent mobility, namely, the mobility necessary to obtain the actual conductivity, assuming that the hot electrons were moving in an applied field with a true drift velocity. Although the mobility thus calculated is imprecise, it is sufficiently accurate to allow simple comparisons.

It is not surprising that the present simple model is inadequate to predict photoconductivity in $Al_2O_3$ and MgO. Because band tailing is not important in crystalline materials, the thermalized electrons in the conduction band make the major contribution to the conductivity. Apparently, some plastic materials are also sufficiently ordered for the same argument to hold. For example, Lee et al.[2] found the photoconductivity of polyethylene to be dependent on the degree of crystallinity, with the more dense and more crystalline polyethylene having a higher photoconductivity. It is also not surprising (see Fig. 2) that some other plastic materials exhibit a prompt conductivity several orders of magnitude greater than the minimum value calculated here. Some measurements of the photoconductivity excited by band-gap wavelengths and x-ray-induced conductivity have been made for a couple of polymers, and the conductivity was found to be the same whether the carriers were generated by 4-eV photons or 500-keV photons.[18] In these cases, thermalized electrons make the dominant contribution to the conductivity; thus, the model presented here is incorrect, predicting a conductivity which is too low. We suggest that much larger conductivities than predicted by the present model indicate that thermalized electrons play a dominant role and that in those cases one expects, and generally observes, that the conductivity is temperature-dependent. Recent measurements of R. H. Barlett (private communication) showed that, indeed, the delayed photoconductivity of a variety of amorphous and plastic materials is temperature dependent, but that for many materials, the prompt conductivity is not temperature-dependent. We infer from the latter that the usual temperature-dependence of the conductivity of thermalized electrons is not observed because the prompt conductivity is dominated by hot electrons. We suggest that at sufficiently short times, all plastic and highly disordered materials exhibit a prompt conductivity that is temperature-independent and that exhibits the minimum value predicted here. What is remarkable is that the contribution of thermalized electrons

appears to be delayed so long for borosilicate glass and some other amorphous or plastic materials. Finally, it is clear from the agreement between our simple model and experiment data that impurities play a relatively minor role in these cases. This is in striking contrast to both their usual dominant role in photoconductivity and to the conclusions of Lee et al.[2]

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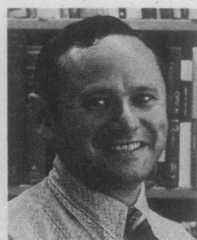
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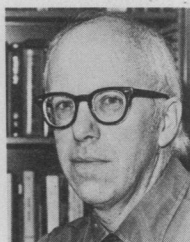
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