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## LETTER TO THE EDITOR

# Frequency scaling of the low-temperature photoconductivity of amorphous silicon

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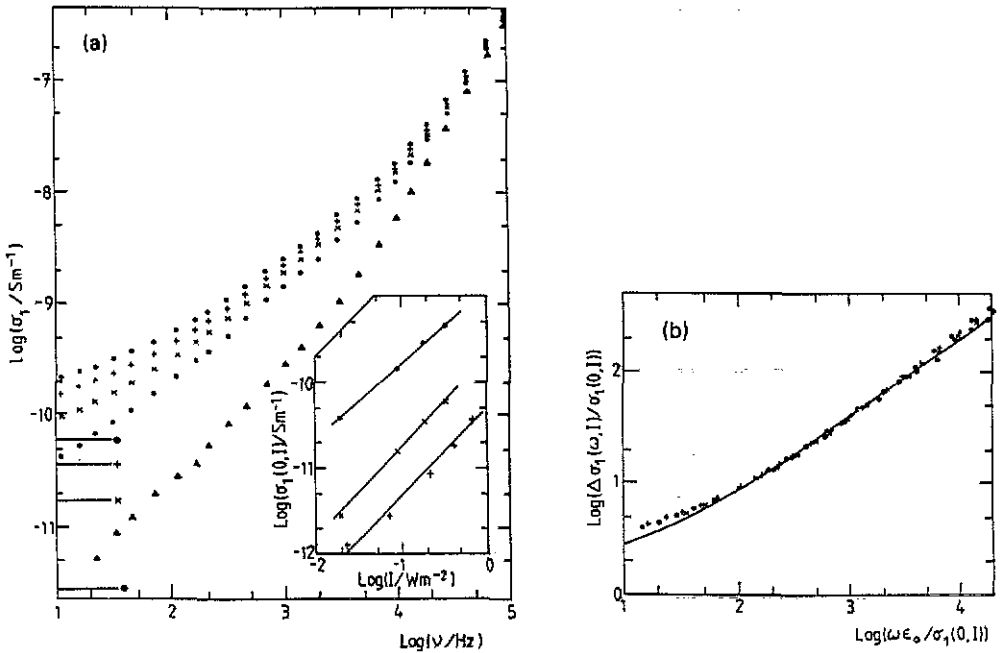
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**Abstract.** The photoconductivity of intrinsic hydrogenated amorphous silicon has been measured between 12 K and 50 K at DC and at audio frequencies. The data for different intensities of illumination normalized to the appropriate DC photoconductivity  $\sigma_1(0)$  follow a universal function of the reduced frequency  $\omega/\sigma_1(0)$ . This *scaling* behaviour suggests strongly that the underlying recombination mechanism involves hopping in band tails. Compensated samples containing significant potential fluctuations have an almost identical response, implying that carriers trapped in wells in the bands do not contribute significantly to the loss. The data are discussed in the light of recent theoretical models.

There have been a number of recent studies of the low-temperature DC photoconductive properties of hydrogenated amorphous silicon (a-Si:H) by, among others, Hoheisel *et al* (1983, 1984) and Spear and Cloude (1987). Hoheisel *et al* interpreted their data as reflecting contributions to the conductivity during thermalization through the extended states whereas Spear and Cloude ascribed the photoconductivity to transport in tail states. Long *et al* (1988) measured the AC photoconductance and photocapacitance of a range of samples for low intensities of illumination, studying particularly the long-term decay of the signal. Recently Johansen *et al* (1989) have reported DC photoconductivity measurements on a wide range of disordered materials. All show similar magnitudes and temperature dependences for the photoconductivity. In a review article Fritzsche (1989) discussed the photoconductivity data, together with drift-mobility measurements by Cloude *et al* (1986) and by his own group.

On the theoretical side, Shklovskii *et al* (1989a) have developed a model (hereafter called the SFB model) in which they successfully explained a range of the available data including the magnitude and temperature dependence of the DC photoconductivity. They attribute the transport process to the hopping of carriers in band tails and assume that the recombination is predominantly geminate in the region experimentally accessible. Subsequently the same authors (Shklovskii *et al* 1989b) elaborated the model to account for the AC loss data of Long *et al*. Searle (1990) criticized the assumption of predominantly geminate recombination in the SFB model, pointing out that the balance of the evidence from luminescence and light-induced spin resonance experiments at typical excitation intensities was in favour of non-geminate recombination. He presented a simpler model, similar in spirit to an earlier analysis of Dunstan and Boulitrop (1984), for the tunnelling recombination at low temperatures, which also gave results in good agreement with experimental data.

The purpose of this letter is to discuss new DC and AC photo-induced loss data for high-quality intrinsic and compensated a-Si:H samples. By AC loss in this context we



**Figure 1.** (a) The frequency-dependent conductivity of an intrinsic a-Si:H sample at 12.5 K.  $\Delta$ : dark,  $\odot$ :  $18 \text{ mW m}^{-2}$ ,  $\times$ :  $89 \text{ mW m}^{-2}$ ,  $+$ :  $180 \text{ mW m}^{-2}$ ,  $\bullet$ :  $340 \text{ mW m}^{-2}$ . The horizontal lines give the appropriate DC conductivities. The superlinear behaviour above 10 kHz is due to series resistance. Inset: the intensity dependence of the DC photoconductance.  $+$ : compensated sample at 12.5 K,  $\times$ : intrinsic sample at 12.5 K,  $\bullet$ : intrinsic sample at 50 K. (b) The data of (a) in scaled form, with the same symbols. Full curve: the prediction of the extended pair approximation with an arbitrarily scaled frequency axis.

mean the response to an alternating applied voltage under steady illumination, not the response to chopped illumination which is associated with the recombination lifetime. These experiments were taken up to higher intensities than in the previously reported measurements of Long *et al* (1988a) so that the relationship between the AC and DC conductivities could be examined. The compensated samples were studied to see whether potential fluctuations, which are expected to be significant in this material (Overhof and Thomas 1989) have any role in trapping carriers and in increasing the response.

The measurements were performed on intrinsic and compensated a-Si:H films prepared by the glow discharge technique and grown to thicknesses of  $\approx 2 \mu\text{m}$ . The compensated samples were prepared from silane pre-mixed with 2.5 vppm of phosphine and diborane. (We are grateful to Professor W E Spear of the University of Dundee for supplying these samples to us.) Measurements were made in sandwich configuration. ITO or tin-oxide layers and aluminium films were used for the bottom and top electrodes, respectively. The samples had  $n^+$  layers at both electrodes to give injecting contacts but the top  $n^+$  layer was etched back after the deposition of the top electrode to eliminate any parallel conduction path. Measurements were made with the sample bonded to a cold station held between 12 K and 50 K. The samples were illuminated using an optical fibre and a HeNe laser (633 nm). The intensity was varied outside the cryostat using

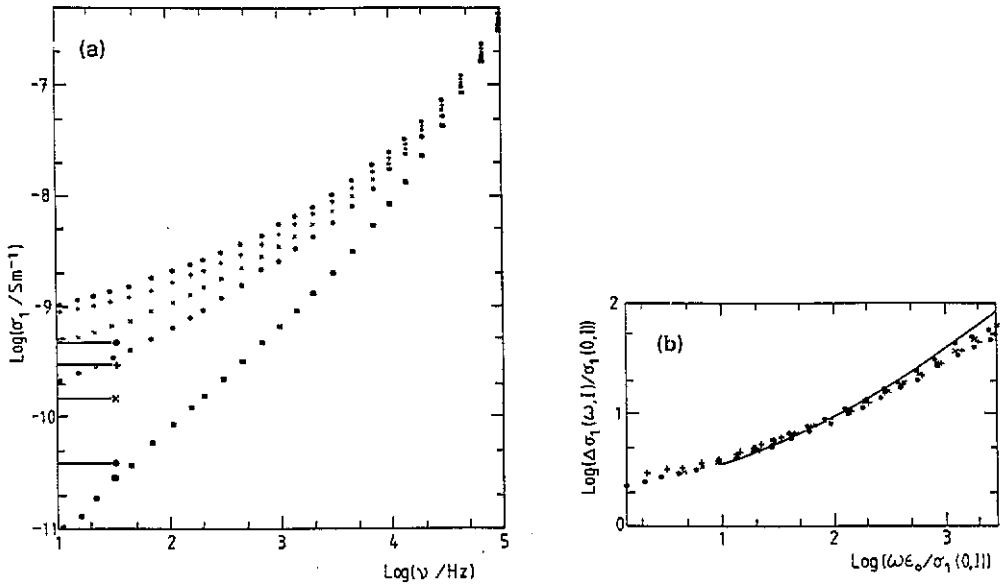
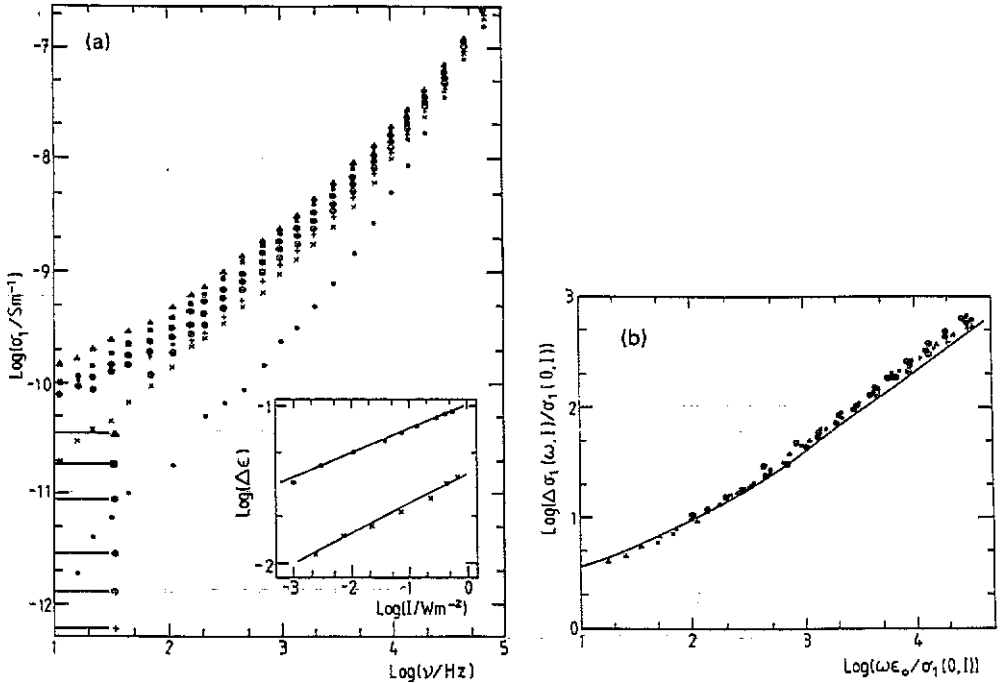


Figure 2. (a) The conductivity of the same sample as in figure 1(a) but at 50 K.  $\square$ : dark. Other intensities as in figure 1(a). (b) The data of figure 2(a) in scaled form, with the same symbols. Full curve: a fit to the data of figure 1(b), transferred at the same scale values.

calibrated neutral density filters. At the end of the experiment a direct calibration of the light intensity falling on the sample was performed. The AC photoconductivity measurements were made between 11 Hz and 100 kHz using either a General Radio 1615A capacitance bridge or a Hewlett-Packard 4274A LCR meter as described in our earlier letter. DC photoconductances were studied using a variable voltage source and a series picoammeter. The dark DC conductances were immeasurably small below 50 K. Upon illumination, however, DC currents did develop; the resulting  $I$ - $V$  characteristics were in general linear between  $\pm 100$  mV suggesting that injection effects can be ignored over this range.

Figures 1(a) and 2(a) show the frequency dependence of the total photoconductivity  $\sigma_1(\omega, I)$  for an intrinsic sample at two temperatures and under varying light intensities,  $I$ , falling on the sample. The equivalent permittivity data for the lower intensities are very similar to those reported in our earlier letter (Long *et al* 1988a). The horizontal lines on the graphs give the corresponding DC photoconductivities  $\sigma_1(0, I)$ . The inset to figure 1(a) shows the relationship between the DC conductivity and the light intensity which is broadly linear ( $\sigma_1 \propto I^\gamma$  with  $\gamma \approx 1$ ) at the lowest temperatures in agreement with previous measurements. At 50 K,  $\gamma$  declines. Note that the temperature dependence of the DC photoconductivity of the intrinsic sample between 12 K and 50 K is much stronger than reported by Johansen *et al* (1989). The data for the compensated material (figure 3(a)) is very similar to that of figure 1(a). The inset to figure 3(a) gives the intensity dependence of the photoinduced response of the compensated sample at 2 kHz. Data for both real and imaginary parts of the permittivity are represented; the intensity dependence of the real part is very similar to that reported in our previous letter for intrinsic material, though the magnitude is higher.

In figures 1(b) and 2(b) we plot the data of figures 1(a) and 2(a) in the form  $\log(\Delta\sigma_1(\omega, I) / \sigma_1(0, I))$  versus  $\log(\omega\epsilon_0 / \sigma_1(0, I))$ . Before scaling in this form, two corrections



**Figure 3.** (a) The conductivity of a compensated sample at 12.5 K. ●: dark, ×: 2.5  $\text{mW m}^{-2}$ , +: 8.5  $\text{mW m}^{-2}$ , ○: 23  $\text{mW m}^{-2}$ , ⊗: 72  $\text{mW m}^{-2}$ , ⊕: 200  $\text{mW m}^{-2}$ , ■: 455  $\text{mW m}^{-2}$ , ▲: 690  $\text{mW m}^{-2}$ . The horizontal lines give the appropriate DC photoconductivities. Inset: the intensity dependence of the change in dielectric constant at 2 kHz. ●: real part (capacitance), ×: imaginary part (conductance). The lines drawn through the data are of gradient 0.15 (real part) and 0.19 (imaginary part). (b) The data of (a) in scaled form, with the same symbols. Full curve: a fit to the data of figure 1(b) transferred at the same scale values.

are made. Firstly, the well known additional contribution to  $\sigma_1$  at high frequencies due to series resistance (see figure 1(a)) is removed, and secondly the dark AC loss from deep states near the Fermi level (Shimakawa *et al* 1987) is subtracted to leave the photoconductance  $\Delta\sigma_1(\omega, I)$ . All the data taken at 12.5 K (figure 1(a)) are reduced in this scaling plot to a common curve. The data taken at 50 K (figure 2(a)) also scale well. However, when compared with the 12.5 K data (the full curve in figure 2(b)) we see that the scaled curves differ, particularly at low frequencies, indicating that the effects of finite temperature on the response do become perceptible below 50 K. The scaled data for the compensated sample are shown in figure 3(b). Once again the full curve in figure 3(b) represents a fit through the data points of figure 1(b), transferred at the same scale values. The magnitudes of the reduced AC losses are remarkably similar both for intrinsic and compensated samples, implying that a common model should be able to account for both sets of data.

This type of scaling behaviour is well known for hopping data (Long *et al* 1988b, Long 1991), where curves taken at different temperatures can be reduced to a common form by plotting against a reduced frequency proportional to  $\omega / \sigma_1(0, T)$ . The physical reason for the success of this transformation arises from a percolation argument. For the infinite DC percolation network there is a critical longest hop with characteristic frequency  $\omega_c$ ; the DC conductivity is proportional to this frequency. If the conductivity

is measured above  $\omega_c$  then only limited percolation clusters will be able to respond and the conductivity will increase with frequency.  $\omega_c$  is generally a strong function of temperature; however when frequencies are expressed in terms of  $\omega_c$ , the percolation aspects of the hopping problem become, to a good approximation, independent of temperature. Hence, the hopping curves can generally be plotted as a universal function of  $\omega/\omega_c$  or  $\omega/\sigma_1(0, T)$ . A number of calculations of the scaling curve in the region of  $\omega_c$  are available and these generally fit the observed temperature-dependent hopping data quite closely (Long 1991).

In this case, the physical problem is somewhat different. Following other authors (Shklovskii *et al* 1989a, Searle 1990) we assume that the injected carriers decay rapidly into the band tails, and that thereafter they hop from site to site, losing energy in the process, before they recombine by tunnelling. We expect the hopping to be a percolative process because of the random distribution of tail states, and hence we anticipate that a critical frequency can be defined to which the DC conductivity will be proportional. Scaling in the form in which we observe it then follows. Reversing the argument, the fact that we observe scaling behaviour is strong evidence that hopping in band tails governs both DC and AC photoconduction. The exact form of the scaling curve is similar to those observed before. In figure 1(b) we also plot the curve predicted by Summerfield and Butcher (1983) using their extended pair approximation. This fits the variable range hopping data well (Long *et al* 1988b, Long 1991) and with a suitable frequency adjustment gives a remarkably good fit to this photoconductivity data, despite the fact that the percolation is taking place in a non-constant density of band tail states. This confirms the hypothesis of Summerfield (1985) (see also Long 1991) that the percolative aspects of these problems dominate any influence of the density of states involved or indeed of the coupling mechanism between states. A detailed calculation will be necessary to show that a scaling curve of the usual form is indeed appropriate for this case.

There is one other possible origin of the scaling behaviour we have observed. In a recent paper, Long (1989) has shown that macroscopic conductivity fluctuations can lead to an AC response reminiscent of that observed in tunnelling systems, with a critical frequency for scaling related to the bulk conductivity relaxation frequency. However, we do not think that fluctuations have an important influence on the data reported here, as the intrinsic and the compensated samples show almost identical photo-induced loss characteristics, even though the intrinsic sample was almost devoid of any fluctuations whereas the compensated sample showed some evidence for them (Lemon 1990).

This observation of scaling behaviour therefore suggests strongly that the basic model of charge percolating in band tails before recombination is basically correct. However, when one comes to compare theory and experiment directly in the light of the observation, problems begin to arise. The existing models suggest correctly that the DC photoconductivity should vary as  $I^\gamma$  with  $\gamma$  close to one. If we approximate the empirical scaling relation in a given limited region of characteristic by a power law:

$$\sigma_1(\omega)/\sigma_1(0) \propto (\omega/\sigma_1(0))^\beta$$

then this implies a relation of the form

$$\alpha_1(\omega) \propto \omega^\beta I^{\gamma(1-\beta)}$$

coupling the frequency and intensity dependences of the AC photoconductivity. The only existing model of the photo-induced AC loss (Shklovskii *et al* 1989b), which uses a pair model to deduce  $\sigma_1(\omega)$ , predicts a frequency dependence of around  $\omega^{0.8}$ , in good

agreement with experiment over limited ranges of frequency, and an intensity dependence of the order of  $I^{0.3}$ , which is generally rather higher than is observed (see insert to figure 3 and Long *et al* 1988a). More seriously, however, this model ascribes the frequency dependence to the usual pair terms involving the distribution of relaxation times within a random population of sites, and the intensity dependence to that governing the changes of electron concentration. The two powers are thus decoupled theoretically. The success of the scaling relation in explaining the data suggests that this distinction is not appropriate, and, therefore, that a new integrated theory will be necessary to account for the data.

In conclusion therefore, we have observed a novel scaling relation in the low-temperature frequency-dependent photoconductivity of amorphous silicon. This strongly supports the model in which the carriers percolate through band tails before recombining. The observation of quite similar behaviour in both intrinsic and compensated samples suggests that any potential fluctuations which might theoretically trap carriers in these samples have only a minor effect. Comparing the scaled data with the existing theoretical models suggests that these will have to be significantly refined before an adequate description of the data emerges.

The other remaining problem in this area, that of distinguishing between the geminate recombination model of SFB and the non-geminate calculation of Searle can also be tackled in principle from the AC loss measurements by looking at the time dependence of the loss, as in our previous paper (Long *et al* 1988a). We hope to address this topic again in a subsequent article.

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