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June 1985

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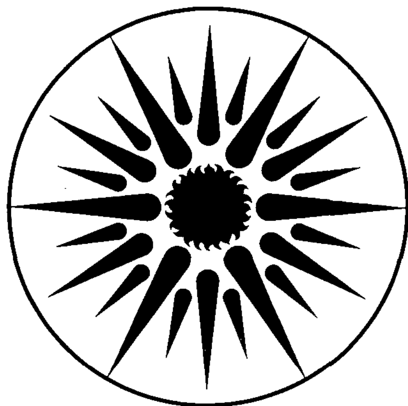

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EVIDENCE FOR ROOM TEMPERATURE TUNNELING RECOMBINATION IN AMORPHOUS SILICON

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Room temperature nonradiative recombination in a-Si:H is directly studied by measuring the thermal expansion of the sample due to the heat released during recombination. We find that the nonradiative lifetime is longest for undoped and shortest for doped material. This is the opposite of the photoconductivity behavior. We conclude that the recombination rates are decoupled from the free carrier densities which is direct evidence that recombination proceeds via tunneling between localized states even at room temperature.

Nonradiative recombination processes in amorphous silicon are studied using a novel technique, photothermal displacement spectroscopy.[1,2] The energy released during nonradiative recombination of photoexcited carriers is converted into heat and the small heat induced thermal expansion of the sample is detected with a Michelson interferometer. The a-Si:H samples were prepared by the glow-discharge method. Undoped and phosphorus doped material has been investigated. The optical excitation source is a Krypton laser operating at 676 nm (400 mW/cm^2, 0.8 cm diameter, 20% power absorption), which is intensity-modulated by an electro-optic modulator at frequencies between 1 and 10^5 Hz. The choice of this wavelength provides a homogeneous illumination of the samples. It also largely reduces the fast heat component associated with the thermalization of carriers towards the bandedges. The interferometer for measuring the thermal expansion (Fig. 1) is operated with a 0.1 mW HeNe-laser (λ = 633 nm), and the sample serves as one of the two mirrors. The position of the reference mirror is adjusted by a piezolectric transducer to maintain a fixed path difference (points A and B in Fig. 1). In this configuration the photodiode signal is directly proportional to the displacement δ ($\delta \ll \lambda$). For a modulated pump beam ($p = p_o e^{j\omega t}$ + c.c) the displacement can be approximated by[2]:

$$\delta = \frac{\xi}{Q\rho} \frac{1}{j\omega} p_o + \text{c.c.} \tag{1}$$

where ξ, Q and ρ denote the thermal expansion coefficient, specific heat and mass density of the sample, respectively. Hence, a $1/\omega$-dependence of the signal amplitude and a phase of -90° establishes the baseline for the dynamic behavior of the photothermal displacement. Any finite recombination effect will show up as a deviation which can be interpreted in a straightforward manner. The feasibility of this approach to investigate nonradiative recombination processes has been demonstrated in a recent study[2]. The overall sensitivity of the interferometer is 3 x 10^{-5} Å for frequencies above 10 kHz.

Typical results for undoped and doped a-Si:H are shown in Fig. 2. In both cases the signal exhibits the $1/\omega$ dependence and a constant phase of -90° as predicted by Eq. (1),

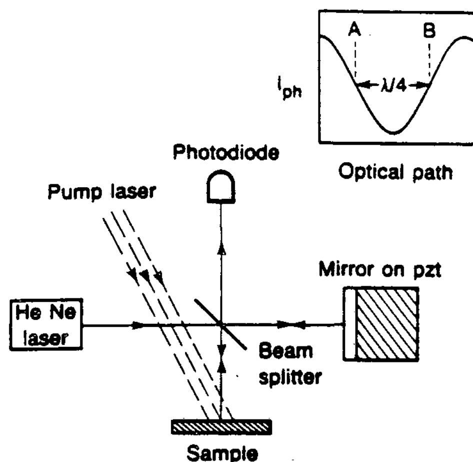

FIGURE 1
Experimental set-up for measuring the photothermal displacement.

except for the very low frequencies where the thermal diffusion length becomes large and heat loss out of the probed area reduces the signal. In addition the undoped sample shows deviations at higher frequencies (> 2 kHz) in both the signal and the phase, which is indicative of a slow nonradiative transition. A fit to an exponential relaxation yields a lifetime on the order of 5 μs. The signal amplitude decreases by nearly 40% with respect to the $1/\omega$ baseline which means that the energy of the nonradiative transition involves at least that portion of the incoming photon energy, i.e., 0.7 eV. The phosphorus doped sample shows no sign of relaxation. We conclude that a large fraction of the incoming photon energy is released in less than 1 μs. Given the experimental accuracy, we estimate this fraction to be $\gtrsim$ 80%. This different behavior was found in several samples and is representative of our undoped and phosphorus doped material.

These results are quite surprising, since it is well known that phosphorus doping sensitizes a-Si:H by enhancing the majority carrier lifetime considerably, resulting in a much higher photoconductivity as compared to undoped material. This effect is believed to be due to the charge state of the dangling bond defect[3]. While this defect is neutral, acting as deep electron trap and recombination center in undoped material, it is negatively charged due to the shift of the Fermi energy E_F in the phosphorus doped case and, hence, incapable of directly capturing electrons. The photocurrent measurements were made on the same samples used in the displacement experiment, and yield the frequency dependence of the majority carrier density. The results (Fig. 3) are clearly in agreement with those reporting long lifetimes ($\simeq$ ms) for doped material and short lifetimes ($\sim\mu$s) for the undoped case. Moreover, the value of the steady-state photoconductivity is also much larger for the doped material (5.3×10^{-4} vs. 3.5×10^{-5} Ω^{-1} cm^{-1}). These results indicate that the carrier density is not related to the recombination rate, but exhibits the reverse behavior: long nonradiative lifetime and short carrier lifetime in the undoped samples, and vice versa in the doped material.

Obviously, these results conflict with the commonly applied Shockley-Read type recombination models. According to this approach, all gap states are essentially isolated from each other, and transitions are allowed only between extended, i.e., conducting, states and localized states. That would imply that the free carrier density (and hence the photocon-

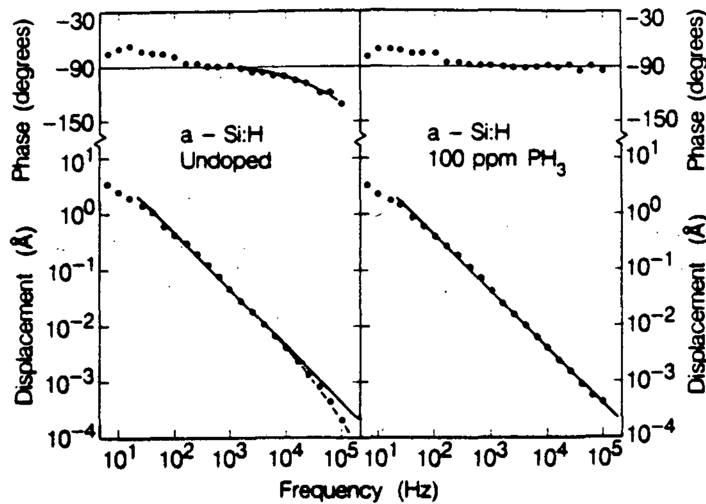

FIGURE 2
Frequency dependence of the photothermal displacement in undoped and doped a-Si:H.

ductivity) is directly related to the recombination rate (i.e., the photothermal signal) which is clearly not the case. A model where carriers recombine by tunneling between localized states in the gap[4], however, is in qualitative agreement with the experimental results. The major reason is that for the tunneling case two restrictions do not apply: First, the energy of the excited electron-hole pair (i.e., the gap energy) can be released in several small steps instead of the maximum of two as prescribed by the Shockley-Read recombination. Second, transitions between deeply localized states which are not allowed in the Shockley-Read case do not involve free carriers, and, in certain cases, may be of little influence on the free carrier density. For the phosphorus doped material, we suggest

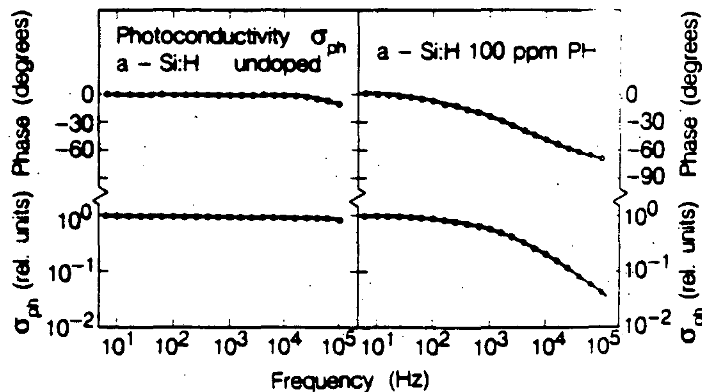

FIGURE 3
Frequency dependence of the photoconductivity in undoped and doped a-Si:H.

that the bottleneck of the recombination is a tunneling transition between localized electrons in the conduction bandtail and the dangling bond. Since the recombination proceeds as a cascade through the localized state distribution, the slowest, hence rate determining step, may involve arbitrary small amounts of energy which are well below the resolution of our experiment. Therefore, the electron population of the bandtail relaxes slowly, which is observed in the photoconductivity. However, by far the largest amount of heat is released instantaneously as indicated by the displacement experiment. In the undoped sample most dangling bonds are in the neutral state. Therefore, capture of electrons is more likely to occur resulting in a fast relaxation of the photoconductivity. In Ref. 4 it was argued that the next transition from the now negatively charged dangling bond to the hole trap above the valence band is much slower in the undoped sample due to the low density of negatively charged dangling bond states. As long as the occupancy of the dangling bond state is not changed appreciably (as we confirmed by the absence of a light induced ESR-signal at room temperature), this transition will not directly influence the electron population. It will, however, show up in the displacement experiment since it involves a considerable amount of energy (on the order of 0.7 eV).

In conclusion, we have shown by a novel technique that recombination in a-Si:H proceeds by a tunneling mechanism rather than by direct capture of free carriers at defects.

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