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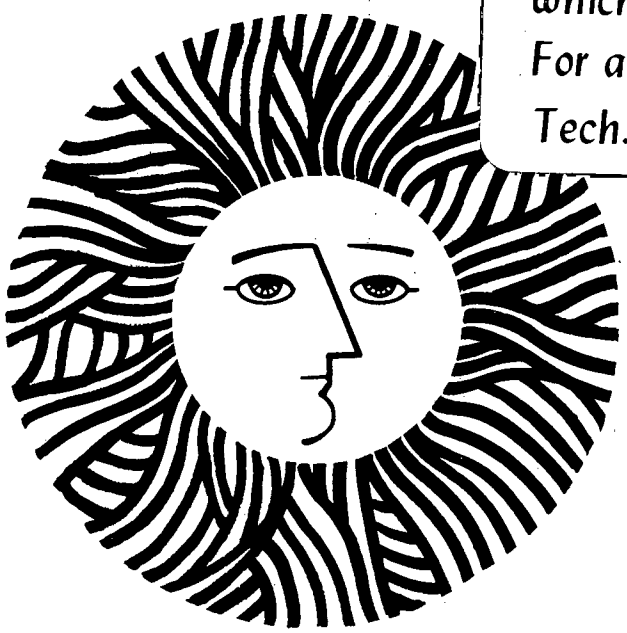

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SUB-GAP AND BAND EDGE OPTICAL ABSORPTION IN a-Si:H BY PHOTOTHERMAL DEFLECTION SPECTROSCOPY

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Using photothermal deflection spectroscopy, we have investigated the optical absorption of various a-Si:H films in the range of 2.1-0.6 eV. An absorption shoulder which depends on deposition conditions and on doping was found and was attributed to dangling bonds. We also observed that the exponential edge broadens with increasing spin density.

INTRODUCTION:

The nature of the optical absorption in amorphous semiconductors at and below the absorption edge is of interest. In particular, the absorption below the band gap is a strong function of defects and impurities since it is not masked by the stronger band-to-band absorption. In the case of hydrogenated amorphous silicon (a-Si:H), since the film thickness is typically one micron thick, conventional methods of reflection and transmission do not reliably measure absorption coefficients (α) below $\sim 50\ cm^{-1}$. And while photoconductivity data have been used to infer low optical absorption, unverified assumptions about the transport properties of the material had to be made. To overcome these limitations, we have recently developed the technique of photothermal deflection spectroscopy [1] (PDS) which enables the direct measurement of low absorption coefficients ($\alpha \ell \sim 10^{-7} - 10^{-8}$). Using this technique, we have measured the sub-gap absorption of various a-Si:H films down to 0.6 eV.

EXPERIMENTAL CONSIDERATIONS:

The physical basis of PDS is that when an intensity-modulated tunable light beam (pump beam) is absorbed, heating will occur. This heating causes a periodic index of refraction gradient in a thin layer adjacent to the sample surface. A second beam (probe beam), propagating through this thin layer, will then experience a periodic deflection which can be quantitatively related to the optical absorption. We have shown [1] that for thermally thin materials, the power-normalized signal S is given by

$$S = A\,[1 - \exp(-\alpha\ell)]$$

where A is a constant which can be determined empirically, and ℓ is the film thickness. For $\alpha\ell \gg 1$, $S = A$ and hence A can be determined.

Experimentally, the output of a 1kW Hg-Xe arc lamp was monochromatized (0.01 eV bandwidth), mechanically chopped, and focussed on the sample which was immersed in CCl_4. The deflection due to absorption was measured with a He-Ne laser (probe) beam whose direction was detected with a position sensor. The output of this sensor was fed into a lock-in amplifier and the pump beam power was monitored with a pyro-electric detector. Interference fringes were averaged out using conventional techniques. Using radiation transfer theory, we have also demonstrated that the PDS signal is highly insensitive to the scattering properties of the films [2]. To insure that the observed PDS signal originated from the a-Si:H film and not from the substrate or the CCl_4, we monitored the phase of the signal. Theory shows that signals from the substrate and from the liquid, should be $+45^{\circ}$ and -135° out of phase with that from the film, respectively. No such phase shifts were observed. We also varied the substrate material and the liquid chemical nature and found that neither affected our results.

The a-Si:H films were undoped r.f. and d.c. glow discharge 1 - 2μ thick. Substrate deposition temperature ranged from 100° - 300°c, and the r.f. power was 2 - 40 W. One phosphorus doped sample (10^{-3} PH_3) was also measured.

RESULTS AND DISCUSSION:

The results are shown in Figures 1 and 2. As the substrate temperature is increased, the shoulder at ∿ 1.3 eV decreases. Then above 230°c the strength of the absorption increases. Furthermore, as the r.f. power goes up, the abosrption shoulder increases in a fairly monotonic fashion. Even though the phosphorus-doped film was deposited at 230°c and 2 W power, it exhibits the largest absorption shoulder (our results from a systematic investigation of single-doped and compensated films will be reported elsewhere [3]). Another characteristic of the absorption spectra is that the slope of the exponential band edge varies with the r.f. power, with the slope being well correlated with the absorption shoulder.

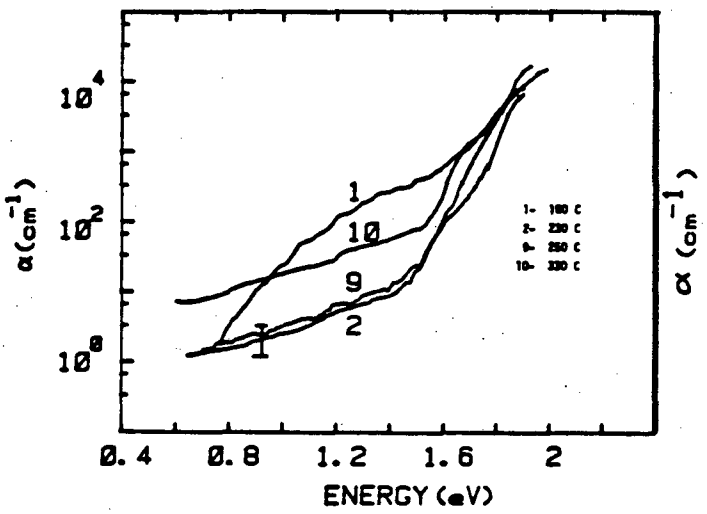

Fig.(1) Dependence of Absorption Shoulder on Substrate Temperature

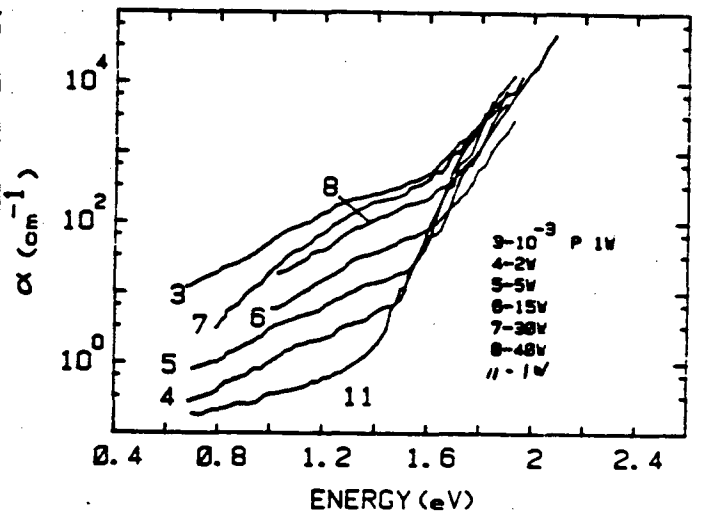

Fig.(2) Dependence of Absorption Shoulder on r.f. power

The absorption in this region can be fitted to the form

$$\alpha = \alpha_o \exp(\hbar\omega/E_o)$$

hence, to attempt to identify the origin of this absorption we plot the equilibrium spin density of the various films (as deduced from ESR measurements) vs. E_o. As can be seen from Figure (3), the correlation is very strong. There are three possible reasons for such a correlation: 1) since the absorption shoulder is attributed to spin related defects [3], such a shoulder may extend into the exponential region. Thus an increase in the spin density would increase the absorption, and in effect flatten the exponential tail; 2) it is believed that the slope of the exponential edge is controlled by disorder-induced fields or strains caused by spin defects; or 3) the spin defects and the disorder are produced under the same deposition conditions.

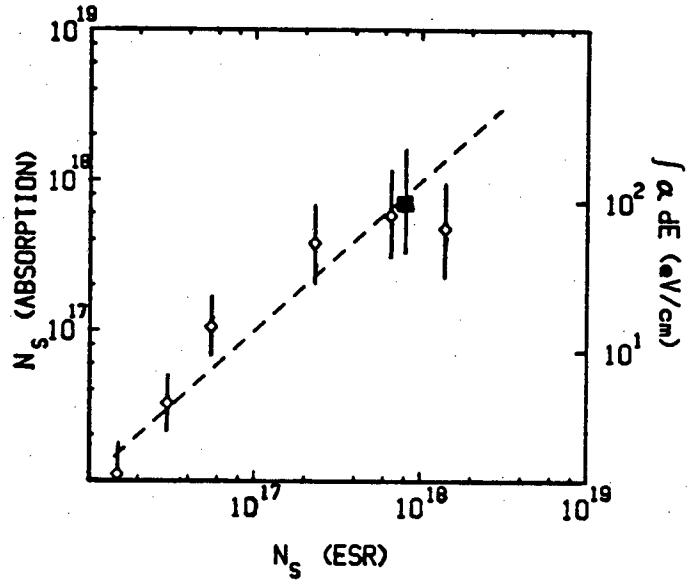

Fig.(3) E_o vs. Equilibrium Spin Density. Black Square is P-doped sample.

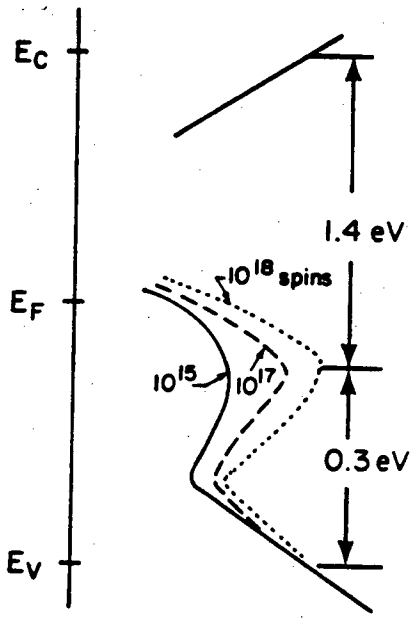

Fig.(4) Schematic Representation of the Effect of Increasing Number of Spins on Density of States

Photoinduced absorption is another way of probing this region of the optical absorption spectrum. It is given by [4]

$$\Delta I \propto t \exp(1 - \alpha)$$

here $\alpha = kT/E_c$ (or E_v), where E_c (E_v) is the slope of the exponential tail of the conduction (valence) band. Hence, one would expect α to be related to the slope of the absorption edge. Preliminary results in our laboratory [5] indicate a good correlation between the photoinduced parameter α and absorption edge slope.

A model to explain our results is schematically presented in Figure (4). As the spin density increases, the valence band defect states increase and flattening of the valence band tail occurs. Evidence for the existence of this structure in

the density of states comes from the strong correlation between the absorption and spin density in undoped, single-doped, and compensated samples [3], field effect measurements [6], and DLTS [7]. This picture is consistent with theoretical and experimental evidence indicating that the valence band is more sensitive to the effects of disorder than the conduction band [8].

Finally, it has been suggested that the luminescence involves transitions from the conduction band to a peak in the density of states. Such a model requires a positive correlation between absorption and luminescence. From our results we find that the absorption and luminescence are not at all correlated [9]. Indeed, one has to conclude that the peak in the density of states quenches the luminescence rather than cuases it.

CONCLUSION:

We summarize our results as follows:

1) A significant absorption shoulder at $\sim$ 1.3 eV is observed which is strongly correlated with spin density.

2) The growth of this absorption feature is accompanied by a broadening of the absorption edge.

3) This broadening provides evidence that the valence band edge broadens as the density of defect grows.

ACKNOWLEDGEMENTS:

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