Generalized model for the optical absorption edge in a-Si:H

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We have reanalyzed the published optical absorption coefficient data for a-Si:H and introduced a divergence temperature, a new concept in the physics of these materials.

An important area of research in disordered substances is the equilibration of such systems. Particularly, the defect kinetics and the effects of structural rearrangements in a-Si:H are topics of current theoretical as well as experimental investigations.[1-3] One reason for this interest is that if a-Si:H is truly far from thermodynamic equilibrium then its physical properties, viz., the electronic properties, will be determined by the deposition process.[2] Hence, it is important to know how close to a metastable equilibrium is a given noncrystalline substance. The low-energy (E) optical absorption, α, in a-Si:H has been studied by Cody et al. as functions of sample treatment, temperature, pressure, etc. They report[1]

$$\alpha(E,T)=\alpha_0\exp(E-E_\infty)/E_0(T,X) \tag{1}$$

for $E \lesssim E_g$, the Tauc optical gap energy. It was correctly argued[1] that for amorphous semiconductors, systematic studies are essential. Furthermore, although both E_g and E_0 are influenced by the noncrystallinity of the specimens, E_0 appears to be directly quantifiable in terms of the disorder. They concluded[1] that there should be a temperature (T) independent component to the band-edge absorption of a noncrystalline material. Operationally it means that when $\alpha(E)$ data are plotted versus E at constant T, or after thermal annealing at different temperatures, then these isotherms would converge to a common $\alpha=\alpha_0$ for $E=E_\infty$. That is, the $\alpha(E)$ data would exhibit an "Urbach focus."[4-7] This convergence was clearly shown[1] in Fig. 1.

It was proposed[1] that the contribution of thermal structural disorder to E_0 are linearly superposable, such that $E_0(T,X)=E_0(T)+E_0(X)$. In the harmonic approximation, $E_0(T,x)=K(\langle U^2\rangle_X+\langle U^2\rangle_T)$ where $\langle U^2\rangle$ is the average of the square of the displacement of the atoms from their equilibrium positions due to the disorder. In the Einstein approximation, Cody et al. obtained

$$E_0(T,X)=\frac{\theta}{\sigma}[(1+X)/2+(e^{\theta/T}-1)^{-1}], \tag{2}$$

where $X=\langle U_X^2\rangle/\langle U\rangle_0^2$ is the contribution due to the zero-point uncertainty. From the above, a linear relationship was predicted between E_g and E_0, i.e., $E_g=E_F-GE_0$, where G is the slope of E_g versus E_0, and E_F is the Abe-Toyazawa parameter.[6]

In this paper we will discuss first an extension of this model. We will relate the structural disorder term $E_0(X)$ explicitly with the annealing temperature T_H. Secondly, we will employ the T_H data from Fig. 1 of Ref. 1 to analyze this extended model.

We assume that annealing produces structural rearrangements. Under that condition the $E_0(T_H)$ associated with annealing at T_H will be proportional to the corresponding (equilibrium) thermal energy, or, $E_0(T_H)\alpha T_H$. Secondly, at T_0 an annealing temperature characteristic
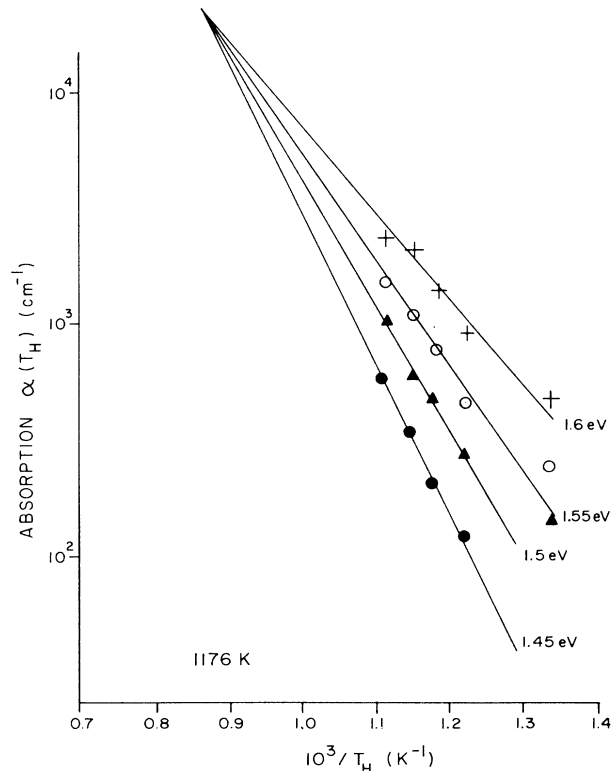

FIG. 1. This is a redrawing of the T_H data from Fig. 1 of Ref. 1. Notice, that (1) the isoenergy $\alpha(T_H)$ behavior is activated, and (2) there is a common focus α_∞ at $T_H=T_0$ of the different "isoenergies."

to the specimen, the disorder energy diverges. Notice T_0 is distinct from the fictive temperature T_f defined in Ref. 8. $E_0(T_H)$ at T_0 will be represented by a simple pole. This choice is not essential in this model but is chosen for analytical convenience. Hence we have

$$E_0(T_H) = \gamma T_H (1 - T_H/T_0)^{-1} \qquad (3)$$

and

$$\alpha(T_H, E) = \alpha_0 \exp[(E - E_\infty)(1 - T_H/T_0)/\gamma T_H] . \qquad (4)$$

The consequences of our model for α will be apparent by expanding Eq. (4). That is,

$$\alpha(E, T_H) = \alpha_0 \exp[-(E - E_\infty)/\gamma T_0]$$
$$\times \exp[(E - E_\infty)/\gamma T_H] \qquad (5)$$

or

$$\alpha(T_H) = \alpha_\infty \exp[(E - E_\infty)/\gamma T_H] = \alpha_\infty \exp(\Delta/T_H) , \qquad (6)$$

where $\alpha_\infty = \alpha_0 \exp(-\Delta/t_0)$ and $\Delta = (E - E_\infty)/\gamma$. Also, from Eqs. (5) and (6)

$$\ln\alpha_0 = \ln\alpha_\infty - \Delta/T_0 . \qquad (7)$$

Notice, there are three parts in the present analysis. These are as follows: $\alpha(T_H)$ is activated in T_{H_1}, i.e., the isoenergy plots of $\ln\alpha_0$ versus T_H^{-1} are linear [Eq. (6)], with a temperature focus at $T_H = T_0$ [Eq. (5)], and $\ln\alpha_0$ is linearly dependent on Δ, the activation energy [Eq. (7)]. Similar behavior is observed in the conductivity data for many thin films in different activated states.[9] This may be indicative of the similarity between the optical and transport behavior reported in these systems.[10] An addi-

tional point of interest is that the behavior represented by Eq. (7) is not sensitive to the exact microscopic model. This is analogous to a similar situation in the transport behavior. Also, the Urbach tail absorption is explainable by a variety of disorder models. This may be why the Urbach behavior is so widely observed in such a wide class of realizations of disorder in physical systems.[11,12] Likewise, the results represented in Eq. (7) might be widely applicable.

Figure 1 shows a replot of the T_H data from Fig. 1 of Ref. 1. These isoenergy data clearly show activated absorption as indicated by Eq. (6). We determine $\alpha_\infty = 1.2 \times 10^6$ cm^{-1}, in close agreement with the value $(1.5 \times 10^6$ cm$^{-1})$ reported in Ref. 1. The characteristic temperature T_0 is estimated to be 1340 K. Recently, Street and co-workers have introduced[2] the glass transition temperature $(T_g \sim 400$ K) of the bonded hydrogen submatrix. Since the α data were read off the Fig. 1, of Ref. 1, the estimate of T_0 is not precise but the order of magnitude T_0 $(\sim 10^3)$ appears to be 3 times that of T_g. As is the case of E_g and E_0, we believe it would be important to study the gradual variation in T_0 due to systematic changes in the specimens. To test the linear dependence of $\ln\alpha_0$ on the activation energy, α_0 and Δ were determined. We find that an excellent straight line fit of $\ln\alpha_0$ versus Δ satisfies the prediction of Eq. (7). We have reanalyzed the $\alpha(\varepsilon)$ data[1] for a-Si:H and introduced a divergence temperature, a new concept in the physics of these materials.[1,2]

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