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Luminescence line shape of amorphous hydrogenated carbon and silicon

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We demonstrate that the experimental luminescence line shapes of amorphous hydrogenated carbon and amorphous hydrogenated silicon are in reasonable agreement with a simplified theoretical model of Dunstan and Boulitrop with no adjustable parameters. Further, the low-energy luminescence intensity is proportional to the joint density of states.

For the last ten years there has been and continues to be a debate over the origin of the luminescence line shape of amorphous hydrogenated silicon (*a*-Si:H).[1,2] Recently, a simple theoretical model for the line shape was proposed by Dunstan and Boulitrop (DB).[2] This model is not only successful in explaining the luminescence line shape of *a*-Si:H, but also its decay kinetics, and its red shift with temperature and excitation energy.[3] In this paper we demonstrate that the luminescence line shape of amorphous hydrogenated carbon (*a*-C:H) is in agreement with the model of DB. We also point out another consequence of the model of DB that relates the luminescence line shape to the joint density of states in both *a*-C:H and *a*-Si:H.

The model of DB starts with the assumption that the luminescence is due to transitions between conduction-(c) and valence-(v) band tail states and that the tail densities of states N, are exponential functions of energy ϵ:

$$N_i(\epsilon)=N_{0i}\exp(-\beta_i\epsilon),\quad i=c,v\ . \tag{1}$$

The probability that a given tail state is the lowest in energy within a volume V is

$$P_{Li}(\epsilon)=\exp[-V_i\beta_i^{-1}N_i(\epsilon)],\quad i=c,v\ . \tag{2}$$

Then the density of tail states that are the lowest in energy within V is

$$P_i(\epsilon)=P_{Li}(\epsilon)N_i(\epsilon),\quad i=c,v\ . \tag{3}$$

Finally, the luminescence intensity at energy E, $I(E)$, is just the convolution over the joint density of deepest valence and conduction-band tail states

$$I(E)\propto\int\int P_c(\epsilon_c)P_v(\epsilon_v)\delta(E-\epsilon_c-\epsilon_v)\,d\epsilon_c\,d\epsilon_v\ . \tag{4}$$

Because the volume V is a function of time, the observed luminescence spectrum is a weighted summation of $I(E)$ with different V. However, we will use a simplified model by choosing average V_c and V_v and inserting them in Eq. (4).

This simplified model of DB has seven unknowns: β_c, β_v, N_{0c}, N_{0v}, V_c, V_v, and E_{0g}, the optical band gap. However because the density of deepest valence-band tail states $P_v(\epsilon)$ is significantly broader than the density of deepest conduction-band tail states $P_c(\epsilon)$, $P_c(\epsilon_c)$ acts approximately like a delta function in Eq. (4). Then Eq. (4) simplifies to

$$I(E)\propto P_v(E)\ . \tag{5}$$

Furthermore, even though the energy of the luminscence line depends on all seven unknowns, the line shape is only a function on one unknown β_v, over a very large range of values of N_{0v} and V_v.[4] Since β_v can be easily measured from the optical-absorption spectrum, the theoretical line shape of the simplified DB model is determined without any unknowns.

Going one step farther, notice that the low-energy side of the luminescence line shape (energies less than the energy at the peak of the line), is largely determined by $N_i(\epsilon)$ since $P_{Li}(\epsilon)$ approaches one. Then for the low-energy luminescence line shape, Eq. (4) simplifies to

$$I(E)\propto\int N_c(\epsilon_c)N_v(\epsilon_v)\delta(E-\epsilon_c-\epsilon_v)\,d\epsilon_c\,d\epsilon_v\ . \tag{6}$$

which is just the joint density of states deep in the band tails. Previously, this joint density of states had been experimentally measured by optical absorption,[5] photoconductivity,[6] photothermal deflection,[7] and photoemission-yield[8] spectroscopies. We suggest that the low-energy luminescence intensity yields the same information.

The ideas presented above can be easily checked against experimental measurements on *a*-C:H and *a*-S:H. In Fig. 1 we have plotted the optical absorbance as a function of energy of an *a*-C:H thin-film plasma deposited from acetylene (C_2H_2).[9] The optical absorbance in Fig. 1 has an exponen-

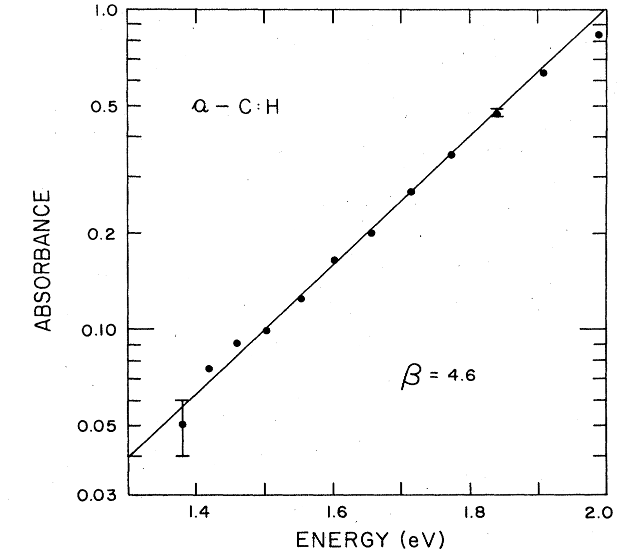

FIG. 1. The theoretical (solid line) and experimental (solid circles) absorbances of an *a*-C:H thin film as a function of energy.

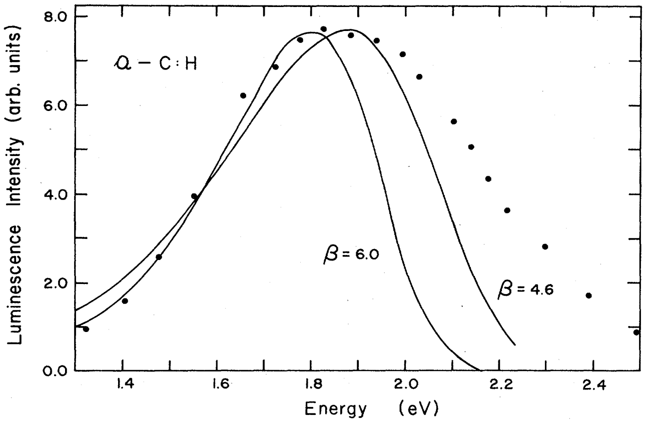

FIG. 2. The theoretical (solid line) and experimental (solid circles) luminescence spectra of a-C:H.

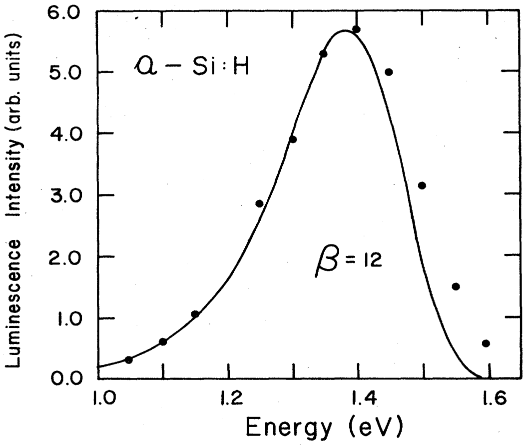

FIG. 3. The theoretical (solid line) and experimental (Ref. 10) (solid circles) luminescence spectra of a-Si:H.

tial tail in agreement with Eq. (1) with $\beta = 4.6$ eV^{-1}. Other a-C:H samples had very similar absorbance curves with β in the range of 4.5–5.0 eV^{-1}. From optical absorption,[5] photoconductivity,[6] photothermal deflection,[7] and photoemission-yield[8] spectroscopies, the values of β for undoped a-Si:H range from 12 to 16 eV^{-1}.

In Fig. 2, the solid circles are the measured luminescence spectrum of the same a-C:H sample whose absorbance was displayed in Fig. 1. The solid lines in Fig. 2 are the theoretical line shapes using Eq. (5) with $\beta_v = 4.6$ eV^{-1} and $\beta_v = 6.0$ eV^{-1}. The theoretical line shape using $\beta_v = 6.0$ eV^{-1} gives the best agreement with experiment in the low-energy region, but the theoretical line shape using $\beta_v = 4.6$ eV^{-1} gives reasonable agreement with experiment over the whole spectrum. Other a-C:H samples gave luminescence spectra whose low-energy line shapes were fitted with values of β_v between 5.0 and 6.0 eV^{-1}. In Fig. 3, the solid circles are the measured luminescence spectrum of undoped a-Si:H taken from Ref. 10, and the solid line is the theoretical line shape with $\beta_v = 12$ eV^{-1}. The agreement between theory and experiment here is very good.

From Figs. 2 and 3, the agreement between theory and experiment is not perfect. First, in this simplified model of DB, we used an average V. By using contributions with different V, the theoretical line shape is somewhat broadened on the high-energy side.[2] Second, the model of DB neglects any thermalization and exciton effects, which are present at short times after excitation.[11] What is encouraging is the approximate agreement between theory and experiment in both a-C:H and a-Si:H using this simplified model with no adjustable parameters. These results give us confidence in the validity of the model of DB and consequently, in our ability to measure the deep-level joint density of states from the low-energy luminescence intensity.

In comparison, the electron-phonon model of Street predicts that the luminescence line shapes should be Gaussian with

$$\sigma = (2 W_p \hbar\omega_0)^{1/2} \; , \tag{7}$$

where W_p is the electron-phonon distortion energy of the self-trapped exciton and $\hbar\omega_0$ is the optical-phonon energy.[12] Fitting the luminescence spectra in Figs. 2 and 3 to a Gaussian gave a good fit with σ(a-Si:H) = 0.15 eV and σ(a-C:H) = 0.40 eV. Using the values of the $k = 0$ optical phonons in single crystal Si and diamond (0.06 eV and 0.16 eV, respectively), we get W_p(a-Si:H) = 0.19 eV and W_p(a-C:H) = 0.53 eV from Eq. (7). Because it is not obvious how to measure W_p independently, it is difficult to compare our experimental luminescence line shapes with the predictions of the electron-phonon model.

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