Subnanosecond radiative and nonradiative processes in a-Si:H

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Radiative and nonradiative processes governing the photoluminescence at early times in *a*-Si:H have been examined with the use of a Ge detector with 250-ps resolution. The broad spectral response of the Ge detector provides a measurement of the total light decay which isolates those rates that affect the net population of short-lived states from relaxation rates of the population packet to lower energy. At 15 K, the initial lifetime is consistently about 8 ns in a variety of samples with a large range of overall quantum efficiencies, indicating an intrinsic source such as the radiative recombination of a localized exciton. The initial lifetime $\tau_0(T)$ falls by about a factor of 5 from 15 to 275 K, and the initial intensity $I_0(T)$ by about a factor of 10 across the same range. The temperature dependences of $\tau_0(T)$ and $I_0(T)$ are compared to those of a number of models in order to probe the underlying physical processes. The results are also compared to a variety of other sub-nanosecond measurements in *a*-Si:H.

I. INTRODUCTION

Photoluminescence (PL) has been used extensively (Refs. 1-4) as a probe of the electronic states and of the dynamics of recombination in amorphous semiconductors. Time-resolved PL experiments have revealed a broad band that shifts to lower energy as a function of delay. At low temperatures the total light decay is characterized by a broad distribution of lifetimes peaking at about 100 μ s, but extending to 10-ns and 100-ms time scales. Recent picosecond experiments⁴ in As_2S_3 glass have found lifetimes of about 2 ns and risetimes less than 30 ps, even for light emitted far below the band edge. There have been some reports⁵ of exponential decay in *a*-Si:H at early times, but the time scale was close to the resolution of the system. In an earlier paper⁶ we described experiments using a detection system with 250-ps resolution which for the first time clearly resolved the fastest radiative processes in a-Si:H. At low temperatures we found no radiative states with lifetimes substantially shorter than 10 ns. The fast PL was characterized by an initial intensity, which monitors the branching ratio into these states, and an initial lifetime which measures the subsequent decay of these states. In this paper we examine these results in more detail and compare the temperature dependence of the initial intensity and lifetime to a number of models in order to probe the underlying mechanisms. The results are also compared to a variety of other subnanosecond measurements in a-Si:H.

II. EXPERIMENTAL TECHNIQUES

Five samples, doped and undoped, and covering a wide range in quantum efficiencies, were used in this study in order to separate intrinsic from nonintrinsic sampledependent properties. The films were prepared by plasma deposition of silane on fused-silica substrates, and were about 1 μ m thick. Films 1–3 were relatively good material in which we estimated the time-integrated quantum efficiency (QE) after pulsed excitation as a few percent. Sample 4 was grown under similar conditions and subsequently annealed at 550 °C for 20 min, which reduced its QE by about an order of magnitude. All four films were anodic, i.e., deposited on grounded substrates. Samples 1, 2, and 4 were undoped, and sample 3 was doped at 100 ppm PH₃ in the gas mixture. The deposition parameters for samples 1-4 are shown in Table I. Samples grown under these conditions in our laboratory consistently contain 14-20 at. % hydrogen, exhibit spin densities below 10^{16} cm⁻³, and have low-temperature optical gaps in the range 1.8-1.85 eV. Thus we expect the optical-absorption coefficient $\alpha \sim 10^4$ cm⁻¹ at 6200 Å. Sample 5 was a low-QE film deposited on smooth fused silica at another laboratory. Its efficiency was about a factor of 2 lower than that of sample 4. A flowing-He-gas cryostat was used to cool the samples.

The samples were excited with a high-power picosecond laser system composed of a synchronously pumped modelocked dye laser and a four-stage excimer-pumped amplifier system. The amplification stages consist of quartzprism dye cells with drilled capillary channels for the flowing dye, originally designed by Bethune.⁷ These cells greatly simplify alignment procedures for this multipleamplifier scheme. Ths pulses from the mode-locked dye laser are passed longitudinally through the capillary channels as the cells are transversely excited with 10 ns uv excimer-laser pulses. The amplified 5-ps pulses deliver 0.5 mJ per pulse at repetition rates from 10 to 200 Hz. The wavelength was fixed at approximately 6200 Å for these experiments.

The luminescence was collected with f/0.75 optics and focused onto a 75-ps-rise-time Ge avalanche photodiode detector (Optoelectronics Model PD 40). The diode output was fed through further stages of amplification, then through a delay line to allow triggering of the electronics, and finally viewed on a sampling scope with a net time resolution of 250 ps. The Ge-diode response varies by only about 30% across the luminescence band. Thus, by inserting a long-pass interference filter (cut on an energy of 1.6 eV) in the collection optics to eliminate scattered

Sample	Substrate	Thickness (µm)	Substrate temperature during deposition (°C)	Gas mixture	Pressure and rf power	Doping
1	roughened fused silica	2.0	250	30 mol% SiH ₄ 68 mol% Ar 2 mol% Kr	0.36 Torr 5 W	undoped
2	same	0.75	250	88 mol % Ar 2 mol % Kr	same	undoped
3	same	1.0	250	$2 \mod \% \operatorname{SIH}_4$ 2 mol % Kr 100 ppm PH ₃	same	100 ppm PH ₃
4	same	1.4	250	10 mol % SiH ₄ 88 mol % Ar 2 mol % Kr	same	undoped

TABLE I. Deposition parameters for samples 1–4. Sample 4 was subsequently annealed at 550 °C for 20 min.

laser light, a good approximation of the total light decay is obtained. Recent subnanosecond experiments using photomultiplier detection^{8,9} have revealed that at very early times radiation is emitted above 1.6 eV. Time-resolved spectra discussed in our previous paper,⁶ however, show that early relaxation processes which bring the spectrum below 1.6 eV are much more rapid than radiative rates,⁶ ensuring that this undetected radiation contributes little to the spectrally integrated intensity. The 1.6-eV filter has the added advantage of blocking any signal from surfacecontamination fluorescence.¹⁰

III. LOW-TEMPERATURE RESULTS

The 250-ps resolution of these measurements allows the fastest radiative processes in a-Si:H, which are approximately 10^{-8} s⁻¹, to be clearly resolved. In fact, however, on the time scale over which the signal drops by an appreciable amount, the decay never becomes perfectly exponential. This is demonstrated in the semilogarithmic plot of the decay in sample 3 shown in Fig. 1, where curvature is apparent by 10-20 ns. In the log-log plot of the same data, however, the pronounced flattening of the slope below 10 ns indicates that the distribution of states with shorter lifetimes is falling precipitously. An approximate determination of the distribution of lifetimes¹¹ is given by $G(t) \sim tI(t)$ which is shown in the inset of Fig. 1. The sharp drop seen below 10 ns is actually resolution limited by this simple calculation of G(t) and may be even steeper. There is an indication of a shoulder in this region of the distribution which may imply a distinct source of the early-time PL, such as a localized exciton, a possibility discussed later. At any rate, there are very few luminescing states with lifetimes shorter than 10 ns. Consequently, an exponential fit at early times is guite good, and for the purpose of further quantitative analysis we define an initial lifetime τ_0 as the slope of the best exponential fit over the first 5 ns. Similarly the initial intensity I_0 is taken as the extrapolation of this exponential fit to zero time.

For all five samples studied we found τ_0 at 15 K to lie

in the range 6–10 ns. We find that I_0 saturates at our highest intensities where we estimate an initial excited-state density of $(1-5) \times 10^{19}$ cm⁻³. The lifetime, however, is not affected by laser intensity, remaining unchanged



FIG. 1. Upper curve: Semilogarithmic plot of PL intensity vs time. Lower curve: Log-log plot of the same data. Inset: Density of rates estimated as $G(t) \sim tI(T)$.

when the intensity is lowered by 2 orders of magnitude into the region where I_0 is approximately linear with intensity. There is no indication of polarization memory in the luminescence.

Since the overall QE of these samples, as well as the initial intensity, vary by more than an order of magnitude, the constant value of τ_0 indicates that the initial decay is due to an intrinsic radiative process unrelated to the defects such as dangling bonds that quench the cw luminescence. The long-time-scale emission in both a-Si:H and chalcogenide glasses has been characterized as radiative tunneling between localized electrons and holes whose rate, $v \sim v_r e^{-2R/R_0}$, falls exponentially with separation. The scale parameter R_0 is the extent of the larger of the electron and hole wave functions, thought to be 10-12 Å, and v_r is the limiting radiative rate expected to be about $10^8 - 10^9$ s⁻¹. This simple picture breaks down at early times where decay rates of the same order as the limiting rate imply separations comparable to R_0 , and a localized exciton description becomes more appropriate. To first approximation, localized exciton emission can be modeled as the radiation from a dipole er_d oscillating at frequency ω and embedded in a medium of dielectric constant ϵ ,

$$v_d = \frac{4}{3}\sqrt{\epsilon} \left[\frac{e^2}{\hbar c}\right] \left[\frac{\omega^3}{c^2}\right] r_d^2.$$

From this expression we calculate $r_d \sim 1.6$ Å, which provides a rough estimate of the wave-function extent of the exciton, indicating a strongly trapped or possibly self-trapped state. Note that this value is considerably smaller than the approximately 10-Å radius suggested by the excitation densities at which saturation occurs in these measurements as well as in longer-time-scale data. The lack of a concomitant change in the decay rate, however, implies that the saturation is not due to the overlap or interaction of these exciton states, but rather to some aspect of the initial excitation process. Although some theoretical work exists on localized excitons in disordered materials,¹² quantitative comparisons with the electronic and lattice energies or with the dynamics of formation and decay are not yet possible.

It should be emphasized that it is the spectrally integrated nature of these measurements using a broad-band detector that allows the decay mechanisms to be isolated from the simultaneous relaxation processes associated with the spectral shift. This explains the apparent discrepancies between the relatively slow decay rates reported here and the distribution of subnanosecond rates observed in other recent experiments^{8,9} employing detectors sensitive only to the high-energy part of the spectrum. We suggest that the fast rates reported in Refs. 8 and 9 are primarily due to the PL spectrum rapidly shifting out of the region of sensitivity of their detectors. A comparison of the results also provides new information on the nature of the relaxation processes. The lack of decay rates on the same time scale as the shift implies that the relaxation mechanisms are dominated by processes that do not separate geminate pairs. Once an exciton has been created, hopping of one carrier away from the other must occur only on time scales which are long compared to 10 ns.



FIG. 2. Measured PL decay at a series of temperatures: \circ -15 K, \blacksquare -85 K, \triangle -127 K, \diamond -153 K, \Box -205 K, and \bullet -250 K.

IV. TEMPERATURE DEPENDENCE

As the temperature is raised both the initial lifetime and the initial intensity decrease. Sample data for film 2 are shown in Fig. 2. Measured values of the initial rate, τ_0^{-1} , and I_0 over the temperature range 15–275 K for samples 1-4, are plotted in Fig. 3, with the initial intensities normalized at 15 K. Data from sample 5 are not included, as the luminescence was too weak to measure accurately at higher temperatures. Although the cw QE in these samples falls orders of magnitudes from 15 K to room temperature, τ_0 and I_0 drop by only factors of 5 and 10, respectively, across this range. We also note from Fig. 3 that τ_0 does not vary with temperature in the same way as I_0 , indicating that different physical processes are involved. This is not unexpected, as τ_0 and I_0 monitor processes occurring on very different time scales. While I_0 is determined by the very fast mechanisms that feed the



FIG. 3. Initial intensities and initial decay rates vs temperature. Solid lines through the $I_0(T)$ data are fits assuming a competing nonradiative process modeled as a MPE into a highly relaxed defect state. Solid-line fits for $\tau_0(T)$ are based on a nonradiative rate $v_{\text{glass}}(T) = v_0 e^{T/T_0}$.

luminescing states, τ_0 monitors the slower decay of this population. In order to probe the identity of the processes that feed and deplete these states, we examine the temperature dependence of I_0 and τ_0 within the framework of various models.

A. Temperature dependence of $I_0(T)$

In crystalline semiconductors where the density of states contains sharp structure, one often observes thermally activated nonradiative rates dominated by a single energy gap ΔE .

$$v_{\rm act}(T) = v_0 e^{-\Delta E/kT}$$

The attempt frequency v_0 is a phonon frequency on the order of 10^{12} s^{-1} . Let us assume in addition the possibility of temperature-independent nonradiative processes that limit the QE even at zero temperature. Since the net non-radiative rate

$$v_{\rm nr} = v_2 + v_{\rm act}(T) \tag{1}$$

is in competition with the rate v_1 which feeds the luminescing states, the branching ratio for luminescence is given by

$$Y_L(T) = \frac{v_1}{v_1 + v_{\rm nr}}$$
.

To make contact with the measured values of $I_0(T)$, we take v_1 to be the rate of the mechanism that populates the states that luminesce on a 10-ns time scale and assume it to be temperature independent. All other competing processes are to be considered "nonradiative," including those that populate long-lived radiative states. Then $I_0(T)$ which monitors the initial population should satisfy the equation

$$\frac{I_0(0)}{I_0(T)} - 1 = \frac{\nu_0}{\nu_1 + \nu_2} e^{-\Delta E/kT} .$$
 (2)

Taking $I_0(0)$ to be the value at 15 K, good fits were found to this expression as shown in the upper half of Fig. 4. It



FIG. 4. $I_0(0)/I_0(T)-1$ and $\tau_0(0)/\tau_0(T)-1$ vs inverse temperature. Solid lines represent fits to Eq. (2).

is interesting to note that very similar behavior has been reported for the initial current peak in picosecond photoconductivity experiments.¹³ The high-temperature intercepts scale with the relative intensities of $I_0(15 \text{ K})$, indicating that v_2 dominates v_1 . This is consistent with the fact that very little of the total time-integrated luminescence is emitted from states with 10-ns lifetimes. With $v_0 \sim 10^{12} \text{ s}^{-1}$, the results place v_2 in the range $10^{10}-10^{11} \text{ s}^{-1}$, a physically reasonable time scale somewhat faster than the resolution of our system. With slightly faster electronics in the detection system, it might be possible to resolve such a process in the risetime of the PL in high-QE samples where this analysis predicts it would be the slowest.

The values for ΔE fall in the range 50±10 meV. These values are consistent with a variety of possible mechanisms including activation out of shallow-band-tail states, exciton breakup, and thermal excitation out of localized states associated with large lattice relaxation. It would be surprising to find such good fits to a single ΔE in the case of activation out of band-tail states unless a clear-cut gap existed in the density of states close to a mobility edge. Such a gap has been suggested by some results,¹⁴ but argued against by others,¹⁵ and has not been predicted to be of this magnitude. It is also difficult to discuss exciton parameters quantitatively because the lack of \vec{k} -space conservation makes the use of the effective-mass approximation somewhat questionable. On the other hand, the formalism developed for crystals to model nonradiative processes involving trapping at point defects with large associated lattice relaxation¹⁶ carries over well to amorphous materials. Multiphonon-emission (MPE) rates are often dominated by soft local modes, a picture likely to be appropriate in a-Si:H. The MPE rate is given by

$$v_{\rm MPE}(T) \sim (T^*)^{-1/2} e^{-E_B/kT^*}, \quad T^* = \frac{\hbar\omega}{2} \coth\left[\frac{\hbar\omega}{2kT}\right],$$
(3)

where E_B is the barrier height at the crossing of the configuration-coordinate curves as shown in Fig. 5 and $\hbar\omega$ is the energy of the dominant local phonons. At high temperatures $v_{\text{MPE}}(T)$ approaches the classical activated rate $v_{\text{act}}(T)$. At low temperatures, however, the rate approaches a constant nonzero value due to the contribution of zero-point vibrations. Thus at high and low temperatures the behavior is identical to that predicted by Eq. (1) where the net nonradiative rate is the sum of a temperature-independent term plus a thermally activated term. The predictions differ only in the curvature of the crossover region, and fits to the two models are equally good. The best fits to the MPE formalism for samples 1-4 are shown in the upper half of Fig. 3.

Although the MPE formalism is mathematically more complex, it has the advantage of being derived from a simple physical model. The dominant phonon-mode energies determined from fits to $I_0(T)$ in samples 1–4 lie in the range 37–39 meV, approximately in the center of the energy range of the single-phonon density of states and well below the peak measured by Raman scattering.¹⁷ This is physically reasonable, as it is fairly common to



CONFIGURATION COORDINATE Q

FIG. 5. Configuration-coordinate energy diagram for trapping at a defect with lattice relaxation. Curve V is the valenceband energy, curve C is the conduction-band energy, and curve D is the defect energy. Competing with v_{MPE} is the rate v_1 populating the luminescing states.

find somewhat softer local modes at point defects that fall in this region of the bulk density of states.¹⁸ Since the MPE rate has not fully reached the classical limit at the highest temperatures, the energy barrier E_B is somewhat larger than determined from fits to Eq. (2), falling in the range 90-150 meV. A specific defect cannot be identified solely on the basis of these results. Nevertheless, one might speculate that since the dangling-bond density is known to correlate with the presence of nonradiative channels in plasma-deposited materials,¹ and is thought to control the initial intensity on a 10-ns time scale in sputtered¹⁹ and plasma-deposited²⁰ materials, it would appear to be an obvious candidate. One must keep in mind, however, that this analysis has defined as competing "nonradiative" processes all channels that do not lead to luminescence on a 10-ns time scale, including those that lead to the longer-lived radiative processes.

The cw QE in a-Si:H has been shown to be consistent with a nonradiative rate

$$v_{\text{glass}}(T) = v_0 e^{T/T_0}$$

with $T_0 \sim 23$ K.²¹ The subscript "glass" refers to the predominance of this functional form in the QE of a variety of chalcogenide glasses. A number of physical models have been proposed as the source of this nonradiative rate. These include spatial tunneling,²² a distribution of activation energies,^{2,14} and phonon-assisted local relaxation at internal surfaces.²³ We have fit $I_0(T)$ to this empirical formula, again assuming that it competes with a temperature-independent rate v_1 that feeds the radiative states. Note that it is not possible to linearize the expression in an equivalent manner to Eq. (2) except in the region $T \gg T_0$. Nonlinear fits to the data yield values of T_0 in the range 50–60 K, somewhat larger than for quenching of the cw luminescence. This is consistent with measurements in our laboratory on 15-ns-to-1-ms time scales where we find that the quenching with temperature becomes steadily more rapid at longer delays,²⁴ corresponding to a continuous drop in T_0 with delay. In other words, the effect of higher temperatures is not only to reduce the subnanosecond branching ratio into the radiative states, but also to shift the distribution of decay rates to shorter times by introducing additional nonradiative recombination paths. The fits of this model to $I_0(T)$ are in any case considerably less satisfactory than the previously discussed models as it fails to reproduce the verv flat region at low temperatures.

B. Temperature dependence of $\tau_0(T)$

The initial rate $\tau_0^{-1}(T)$ is a direct measure of the sum of the radiative and nonradiative decay rates of the population in the luminescing states. We examine the same models discussed for the subnanosecond branching ratios monitored by $I_0(T)$, but there is no reason to expect that the same mechanisms will be operative in the picosecond and nanosecond regimes. Within the various models, the mathematical expressions for $\tau_0(T)$ are the same as those derived for $I_0(T)$, although in this case v_1 is the radiative rate. Fits to Eq. (2) assuming a thermally activated nonradiative rate given by $v_{nr} = v_2 + v_{act}(T)$ are shown in the lower half of Fig. 4. The energy gaps thus obtained are 27 ± 7 meV. There is a serious problem with this approach, however: The high-temperature intercepts are not consistent with the rest of the model. Since we know the net rate at low temperatures, $v_1 + v_2 \sim 10^8 \text{ s}^{-1}$, the values of the intercepts given by $v_0(v_1 + v_2)^{-1}$ should be $10^3 - 10^4$ instead of the 11 ± 5 determined from these fits. A similar problem exists for the MPE formalism, indicating that different mechanisms are indeed responsible for the decay of the luminescing states than for the initial branching ratio.

A more satisfactory representation of the data is obtained assuming the empirical nonradiative rate $v_{\text{glass}}(T)$ discussed earlier. The best fits of this formalism to the data from samples 1-4 are displayed as solid lines in the lower half of Fig. 3 and yield radiative rates v_1 $\sim 1.0 \pm 0.2 \times 10^8$ s⁻¹, $\nu_0 \sim 0.27 \pm 0.7 \times 10^8$ s⁻¹, nonradiative prefactors and scale temperatures $T_0 = 95 \pm 5$ K. The careful reader may note that straightline fits, i.e., fits with a negligible radiative component, are only slightly inferior. Nevertheless, the consistency of the small curvature coupled with the invariance of the low-temperautre limiting values in samples with a wide range in QE strongly support the presence of a dominant radiative rate at low temperatures. As was also reported for As_2S_3 ,⁴ the scale temperatures are a factor of 3-4 times larger than those determined from the temperature dependence of the cw intensity. As discussed for $I_0(T)$, this weakening of the temperature dependence at early times is consistent with trends we observe on longer time scales. Within the framework of this formalism, however, the physical significance of a large T_0 at early times is not clear for either of the two materials.

Recent photoinduced-absorption (PA) experiments in plasma-deposited *a*-Si:H reveal rather different dynamics in the decay of the PA. Namely, decay times of 50 ps at 80 K increasing to 1 ns at room temperature are noted in the PA signals,²⁵ as opposed to the much slower PL decay

times reported here which decrease as the temperature is raised. We propose that different excited-state populations are responsible for the two processes. Further evidence for this proposition comes from recent experiments in which nanosecond PA measurements have been correlated with electron-spin-resonance (ESR) data measuring the spin density N_s .²⁶ In contrast to the PL data shown here, where the early time branching ratio into the luminescing states is greatly reduced in the presence of high spin density, nanosecond PA signals at low temperatures are insensitive to N_s . We suggest that PL arises primarily from the subset of carriers trapped as geminate pairs, while PA may be a probe of the separated carriers, or equivalently of both populations but with different absorption cross sections. There are, in fact, strong indications that PA experiments are sensitive only to holes²⁶ and that trapping as geminate pairs acts as a quenching mech-anism for the PA signal.²⁵ On the other hand, it must be noted that the proposition of different populations is at odds with the assertions that both the luminescing¹ and absorbing²⁶ states are excited with greater than 50% QE at low temperature. The PL QE estimates, however, come from cw experiments where we cannot rule out a nongeminate contribution. In our own pulsed experiments we estimate the time-integrated QE in good-quality, low-spindensity samples to be only a few percent, as in the films studied here. The authors of Ref. 26 who argue in favor of a unique population for PL and PA also point to the similar thermal quenching of the two signals. We suggest that the competing subnanosecond quenching mechanism observed here in PL and modeled as a MPE from extended states into highly relaxed defect states, could easily deplete both the geminate and nongeminate populations. This suggestion is further supported by the identical quenching behavior of $I_0(T)$ reported here and of the initial current peak observed in picosecond photoconductivity experiments¹³ in which only separated carriers contribute to the signal.

V. SUMMARY

A Ge avalanche photodiode detection system with 250ps resolution has been used to resolve the fastest radiative processes in a-Si:H. A study of five samples from two different sources with a considerable range in QE has revealed that at 15 K the initial lifetime of the luminescing states is consistently 8 ± 2 ns. We surmise that the decay mechanism is intrinsic to the a-Si network, and possibly best described as the radiative decay of a localized exciton. Both the initial intensity $I_0(T)$ and the initial lifetime $\tau_0(T)$ decrease with temperature and have been measured from 15 to 275 K. $I_0(T)$ provides information on the very fast processes that determine the branching ratio into the luminescing states. The temperature dependence of $I_0(T)$ is modeled equally well by competition between the process that feeds the luminescing states, and (a) a classical thermally activated process coupled with a temperatureindependent term, or (b) a MPE process into a localized state with substantial lattice relaxation. Fits to the physically realistic MPE model yield values of the energy barrier, $E_B \sim 90-150$ meV, and energies of the active phonon modes, $\hbar\omega \sim 37 - 39$ meV.

These models lack self-consistency when applied to the observed temperature dependence of $\tau_0(T)$ indicating that different quenching mechanisms are operative in the picosecond and nanosecond regimes. The $\tau_0(T)$ results are fit instead to the empirical formula $\tau_0^{-1}(T) = v_1 + v_0 e^{T/T_0}$ found to describe the quenching of the cw luminescence. Values determined for T_0 are somewhat larger than those in the cw case. The PL data do not exhibit the same dynamics observed in PA, possibly because a complementary set of states are being measured.

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