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Excitation and temperature quenching of Er-induced luminescence in *a*-Si:H(Er)

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Photoluminescence (PL) and light absorption of Er-doped amorphous hydrogenated silicon are studied in the temperature range 77–300 K. In the linear pumping regime the intensity of the Er-induced luminescence decreases by a factor of about 15 in this temperature range. Frequency resolved spectroscopy shows that in the same temperature range the lifetime of the excited erbium ions in the amorphous matrix decreases by a factor of 2.5 only. Excitation spectroscopy proves that the primary step of the excitation mechanism is the absorption and free carrier generation in the amorphous matrix. The emission is excited effectively by subgap light in the range of the Urbach edge and even in the range of defect absorption. Based on these experimental findings we propose a defect-related Auger excitation (DRAE) mechanism of erbium luminescence. Probabilities of the DRAE and the competing radiative defect recombination processes ($D^0+e\rightarrow D^-$) are calculated. It is shown that the probability of the DRAE process is larger by an order of magnitude. In this model the temperature quenching of the erbium luminescence observed above 200 K results from the competition of the DRAE and multiphonon nonradiative recombination at the defects. [S0163-1829(97)08036-3]

I. INTRODUCTION

Photoluminescence and electroluminescence of rare earth (RE) elements that are embedded in a solid-state matrix are of great interest because of possible optoelectronic applications.[1] RE ions exhibit luminescence arising from transitions in the inner $4f$ shell at photon energies that are almost independent of the host matrix. Of particular interest is Er, which in its 3+ state emits at a wavelength of about 1.54 μm, very close to the minimum absorption of optical fibers. This emission arises from transitions from the excited $^4I_{13/2}$ to the ground-state $^4I_{15/2}$. It is well known that f-f transitions are dipole forbidden but become partially allowed when RE ions are incorporated in a solid matrix due to an admixture of other orbital momentum states to the f-wave functions. The properties of Er-doped semiconductors have been quite extensively studied both theoretically[1,2] and experimentally.[1,3–8] Silicon, being the base material of semiconductor electronics, has attracted particular interest led by the hope to realize optoelectronic applications of this indirect semiconductor. Recently, LED's using Er-doped crystalline Si have been prepared that operate at 300 K.[9,10] The main observations may be summarized as follows: Er^{3+} emission from crystalline silicon reveals pronounced temperature quenching.[3,4,11] Oxygen doping enhances the emission intensity.[3,7] It turned out that the temperature quenching is a strongly limiting factor for device applications. It has been argued that this effect may arise from both an enhancement of nonradiative deexcitation of erbium ions and a decrease of excitation rate.[10–12]

Photoluminescence of Er-doped amorphous silicon, *a*-Si(Er), was first reported in Refs. 13 and 14. Recently we found that efficient photoluminescence can be obtained in Er-doped amorphous hydrogenated silicon, *a*-Si:H(Er), prepared by magnetron-assisted silane decomposition.[15] Using this technique we prepared an electroluminescent device with pronounced emission at room temperature.[16] In this paper we report on a more detailed investigation of the influence of the amorphous matrix on the excitation and quenching mechanism of the Er-induced emission.

II. EXPERIMENTAL PROCEDURE

The samples of *a*-Si:H(Er) used in this study were prepared by magnetron-assisted silane decomposition technique, where gas mixtures of Ar, $\mathrm{SiH_4}$, and $\mathrm{H_2}$ are used as sputtering gases. This method has been described in detail elsewhere.[17] The silicon targets were partially covered by small metallic platelets of erbium and the Er concentration in the films was varied by variation of the Er coverage. The films were deposited at a substrate temperature of 300 °C on crystalline silicon and fused silica substrates that had been roughened in order to avoid interference fringes. The film thickness amounted to 1 μm. The hydrogen content as determined by IR spectroscopy from the absorption band at 630 cm^{-1} was in the range 6–12 at. %, the major part being incorporated in monohydride configuration. The oxygen content was not varied deliberately. However, according to secondary-ion-mass-spectroscopy measurements our samples contained oxygen in a concentration of 10^{20} cm^{-3}due to contamination. Both constant photocurrent measurements and photothermal deflection spectroscopy were employed to

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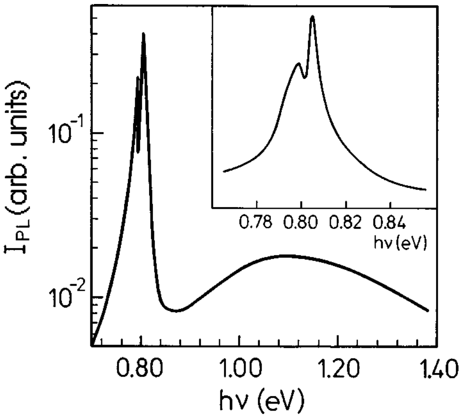

FIG. 1. Photoluminescence spectra of *a*-Si:H(Er) at 77 K (647 nm, 100 mW/ cm^2). Inset: Er^{3+} luminescence at 300 K (647 nm, 350 mW/ cm^2).

study the absorption edge. Optical transmission spectroscopy was used to obtain the absolute scale for the absorption constant. From the subgap absorption we inferred defect densities in the range $5\times10^{17}-10^{18}$ cm^{-3}, applying the procedure as described in Ref. 18. The concentration and the spatial distribution of the incorporated Er was controlled by Rutherford backscattering. We achieved Er concentrations of up to 10^{21} cm^{-3} homogeneously distributed in films of 1 μm thickness. The photoluminescence (PL) was excited by a Krypton laser (647 and 633 nm), an Argon laser (488 nm), or a tunable Ti-sapphire laser (700–1000 nm). The emission was dispersed in a 0.85 m double monochromator and detected by a cooled Ge detector. The spectra were corrected for the system response. The kinetics were studied by frequency-resolved spectroscopy (FRS). For this we modulated the exciting light at frequencies in the range 1–20 kHz and recorded the in-phase signal by lock-in technique. We used a fast Ge detector (North Coast JO 870F) and checked that the response time of the system was less than 2 μs.

III. EXPERIMENTAL RESULTS

The PL spectra measured at 77 K exhibit emission of both Er^{3+} centers and the amorphous Si matrix (Fig. 1). The latter is seen as a broad band at 0.9–1.3 eV. It is widely accepted that this band arises from radiative tunneling between localized band-tail states. Due to the high defect density in our samples of about 10^{18} cm^{-3} the intensity of this intrinsic emission is low and shifted somewhat to lower energy. However, there is no indication of defect-related emission, which in *a*-Si:H of low quality can be observed at 0.8–0.9 eV. In our previous study[15] this emission band formed a broad background of the Er^{3+} photoluminescence, which might be due to the fact that those measurements had been performed at much higher excitation intensities.

The spectrum in Fig. 1 is dominated by strong Er-induced emission at 0.8 eV. At 300 K the emission from the *a*-Si:H matrix is suppressed by temperature quenching and the remaining signal is entirely due to Er^{3+} ions. This emission is shown in the inset on an extended scale. The shape of this band resembles the Er^{3+} band observed at low temperature in crystalline silicon doped with Er and O.[3] It consists of two narrow lines at 0.806 and 0.798 eV that are considered to arise from crystal-field splitting of the ground state ($^4I_{15/2}$) of the Er ions. The emission intensity $I_{\rm PL}$ strongly depends on the concentration of the incorporated Er (Fig. 2). The results in Fig. 2 suggest that there is an optimum around 10^{20} cm^{-3}. The decrease at higher concentrations could arise from cluster formation or, if oxygen played a similar role in *a*-Si:H as in crystalline silicon, from an unsufficient concentration of oxygen in the films. In fact, the oxygen concentration in our specimens is in the order of 10^{20} cm^{-3}. However, as will be shown below, according to our model the limiting factor could as well be the defect density. This model predicts that the concentration of optically active Er ions should not exceed the concentration of defects.

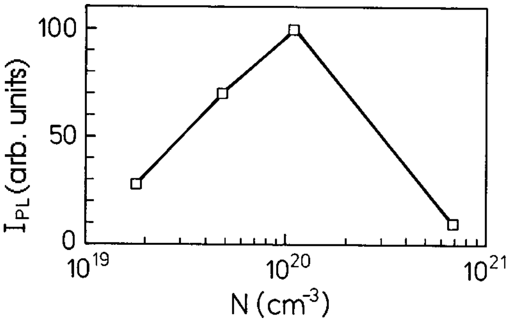

FIG. 2. Dependence of emission intensity $I_{\rm PL}$ on the concentration of incorporated Er ions N. Excitation: 488 nm, 100 W/cm^2, 300 K.

The above result once again demonstrates that the energy position of the Er-induced photoluminescence is only slightly influenced by matrix effects. However, the matrix may have a pronounced influence on the excitation mechanism of the radiative centers. We arrive at this conclusion by comparing the dependence of PL intensity of the Er-induced emission on the photon energy of the exciting light with the absorption edge of this amorphous film. For this experiment it is important to use low excitation power to ensure that the photoluminescence is not saturated. Figure 3 displays the dependence of $I_{\rm PL}$ on the pumping power $I_{\rm ex}$. Following a power law with an exponent of 0.85 the behavior deviates somewhat from the linear dependence expected at the low values of $I_{\rm ex}$. This interesting result will be discussed below in Sec. IV. For the following the important point is that at the excitation power used we are far from the saturation regime. In previous work[15] we pumped with an argon laser which led to saturation due to the much higher values of $I_{\rm ex}$. Figure 4 demonstrates that at constant excitation power $I_{\rm PL}$ nicely follows the energy dependence of the absorption edge of the amorphous matrix. In the region of the Urbach edge ($h\nu>1.5$ eV) carriers are excited between the band tails or from the band tails into extended states above the mobility edges. It is interesting to notice that the parallel dependence of $I_{\rm PL}$ and the absorption coefficient α appears to be valid even at lower photon energies $h\nu<1.5$ eV, where, according to the shape of the absorption-edge absorption from deep defect states prevails and therefore only one mobile carrier per absorbed photon is generated. This behavior shows convincingly that the Er^{3+} centers are not excited by a direct resonant absorption process. Thus the initial step in the excitation mechanism is the absorption in the matrix, which leads to the creation of mobile charge carriers.

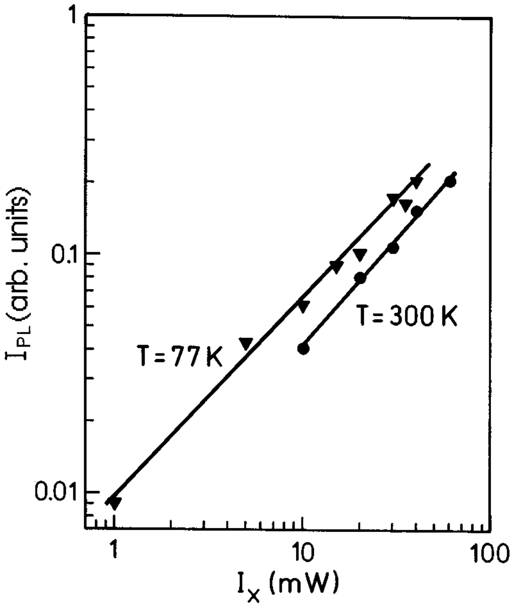

FIG. 3. Emission intensity I_{PL} as a function of the excitation power I_{ex} at 77 and 300 K (647 nm). The scale for the intensity is relative and different for the two temperatures.

The temperature dependence of the intensity of the erbium-induced photoluminescence I_{PL} in a-Si:H(Er) shown in Fig. 5 (curve a) suggests two different regimes. At low temperature I_{PL} decreases only slightly. Only above about 200 K is the photoluminescence strongly quenched and the slope of this curve suggests an activation energy of about 250 meV. Curve b shows for comparison the temperature dependence of the intrinsic emission band of the amorphous matrix (Fig. 5, curve b). The two bands behave quite differently, which suggests that the quenching mechanisms of the two bands is different. It is important to notice that these measurements have been performed using low pumping power such that the photoluminescence intensity does not saturate (see Fig. 3). In the saturation regime the temperature dependence is much less pronounced, as has been reported in Ref. 15. This different behavior in the two regimes will be discussed in more detail in Sec. IV.

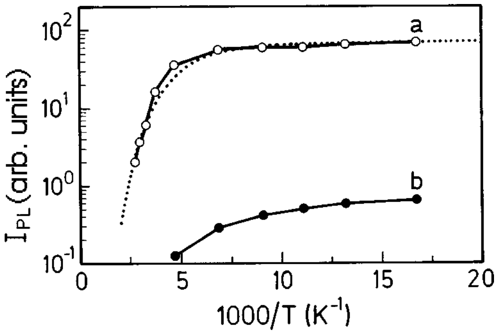

FIG. 5. Temperature dependence of the photoluminescence intensity I_{PL} of the 1.54-μm emission (curve a) and of the intensity of the intrinsic luminescence band at 1.3 eV (curve b) in a-Si:H(Er). Excitation: 647 nm, 100 mW/ cm^2. The dashed line represents the temperature dependence of reciprocal probability of multiphonon capture of an electron by the D^0 center, w_{mp}^{-1} [Eq. (10); see Sec. IV].

The pronounced decrease of I_{PL} with increasing temperature in Fig. 5 can be explained by an enhancement of the nonradiative deexcitation rate of erbium ions or by a decrease of the excitation rate. To distinguish between these two possibilities we investigated the decay rate of the luminescence of the erbium ions. We used FRS to study the kinetics at 77 and 300 K of the a-Si:H(Er) sample. This method has quite frequently been used to determine lifetime spectra and has been shown to be equivalent to time-resolved experiments.[19] Figure 6 shows the in-phase signal as a function of the modulation frequency. These measurements have been performed at pumping powers of 10 mW at 77 K and of 100 mW at 300 K. It was checked that the pumping power had no influence on the frequency dependence of the signal. The lifetime can be defined by the decrease of the signal to half its initial value at the frequency $f_{1/2}$ by $\tau = 1/2\pi f_{1/2}$. We find 20 μs at 77 K and 8 μs at 300 K, respectively. This small value of the lifetime may be the reason why we did not observe saturation as a function of I_{ex} in Fig. 3 when using the Krypton laser for excitation. Between 77 and 300 K the lifetime decreases by a factor of 2.5. The PL intensity, on the other hand, decreases in the same temperature range by a factor of about 15 (Fig. 5). This suggests that the temperature quenching arises predominantly from a decrease of the excitation rate and that the enhancement of the nonradiative deexcitation of erbium ions is less important. Shin *et al.*[20] arrived at the opposite conclusion in the case of a-Si:H(Er),

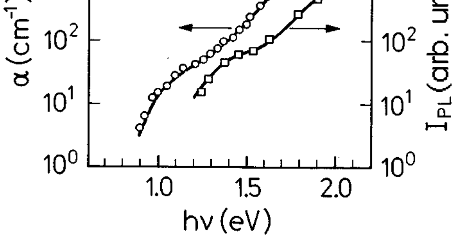

FIG. 4. Photoluminescence intensity I_{PL} and absorption constant, α, of a-Si:H(Er) as a function of photon energy of the exciting light.

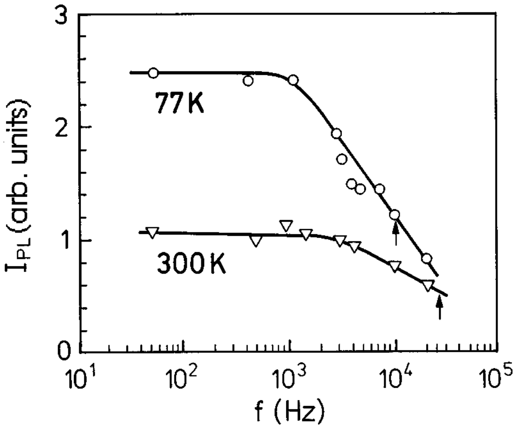

FIG. 6. Real part of photoluminescence intensity as a function of the frequency of the exciting light at 77 and 300 K. Excitation: 10 mW (77 K), 100 mW (300 K).

where the doping had been achieved by ion implantation. They found lifetimes of the same magnitude for *a*-Si:H(Er) and a quite similar temperature dependence. However, by extrapolating their lifetime data into the high-temperature range, it is concluded that the decrease of the lifetime follows that of the PL intensity. A reason for this different behavior could be the different preparation of the samples.

IV. DISCUSSION: THE EXCITATION MECHANISM

The energy position of deep f-electron multiple states lying below the valence states of the semiconductor (by ~ 10 eV) is practically independent of the host matrix due to screening of the f electrons by the external $5s^2 5p^6$ electrons. The energy transfer from electrons of the conduction band to the f-multiple states is produced by Coulomb interaction. In principle, there are three possible processes of electronic excitation of the f states: impact excitation by mobile carriers, Auger excitation in the recombination process of band electron-hole pairs, and an Auger process in which the electron (or hole) is captured in a localized state in the forbidden gap.

The impact excitation where initial and final electron states belong to the same band is efficient only if there is a large number of mobile electrons with high energy. In the case of photoexcitation the fast energy relaxation with a characteristic lifetime $\tau_E \sim 10^{-12}$ s renders this process unlikely. Even in the case of electroluminescence impact ionization is effective only at high electric fields and in materials with large mean free path of mobile carriers. In an amorphous matrix where the mean free path is small, this process is not efficient.

Auger excitation by transferring the energy released by carrier recombination to the f shell seems far more probable. In amorphous silicon we can distinguish two possible channels. The first one is related with transitions between the band tails of conduction and valence bands. The corresponding radiative transitions lead to a broad band of intrinsic luminescence centered around 1.1–1.3 eV. We can also expect a nonradiative transition between the tails of the conduction and valence band with the energy transferred by Coulomb interaction to an f-electron from the ground-state ${}^4I_{15/2}$ to the second-excited-state ${}^4I_{11/2}$. Because the energy of this transition in the f shell is 1.26 eV this process can be considered as a resonance band-to-band Auger excitation. If this process played an important role in amorphous silicon, one should observe similar thermal quenching for the Er^{3+} and the intrinsic luminescence as both bands are produced by electron-hole pairs thermalized in the band tails. However, as shown in Fig. 5, temperature quenching of erbium luminescence starts only at $T \geqslant 200$ K, where the intrinsic luminescence is already absent.

The second Auger process is associated with the capture of an electron from the conduction-band tail by a neutral defect state D^0 transforming it into D^-. These defect centers are attributed to dangling bonds, which can exist in differently charged states (D^+, D^0, and D^-). The temperature dependence of the conductivity revealed that our *a*-Si:H(Er) samples are slightly *n* type with the Fermi level located 0.3 eV above midgap. We assume that in equilibrium practically all defects are in the D^- state. The subgap-absorption measurements have shown that the introduction of erbium into the amorphous matrix is accompanied by a very high concentration of defects. It is therefore reasonable that defects exist in close vicinity to the optically active erbium ions. When the electron-hole pairs are generated by light absorption, they recombine predominantly by nonradiative processes via dangling bonds. Capture of holes by D^- centers, transforming them into D^0, is followed by capture of electrons from the conduction-band tail. We propose that an effective, nearly resonant excitation of f electrons occurs by the electron capture in D^0 states due to an Auger process. This process would be close to resonant because the energy of between 0.8 and 0.9 eV of the $D^0 + e \rightarrow D^-$ is close to the transition energy ${}^4I_{15/2} \rightarrow {}^4I_{13/2}$ in the f-shell transition (0.8 eV). We introduced this process as an explanation of our results on electroluminescence in *a*-Si:H(Er) as DRAE.[15,21] A configuration diagram displaying the various $D^0 + e \rightarrow D^-$ transitions is presented in Fig. 7: radiative capture (1), the DRAE nonradiative transition (2), and a multiphonon nonradiative transition (3). The diagram represents adiabatic potentials corresponding to initial (i, $D^0 + e$) and final (f, D^-) states of the system of an electron and the defect D, taking into account the electron-phonon interaction (local vibrations of the defect). The f states of the Er^{3+} ion are also shown. The dashed line indicates the adiabatic potential for the intermediate (virtual) state of the DRAE process when the electron captured by the defect excites the erbium ion. As stated above, the DRAE process is nearly resonant and its probability should depend only slightly on temperature. It should be noted that precise resonance is not necessary since

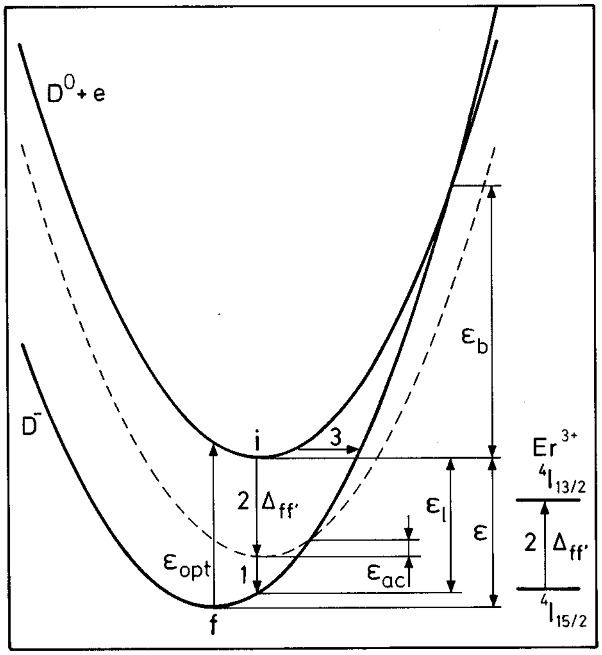

FIG. 7. Configuration coordinate diagram for electron capture by D^0 states ($D^0 + e \rightarrow D^-$). (1) Radiative transition (0.8–0.9 eV). (2) Defect-related Auger excitation of Er^{3+}ions. Both electron capture and excitation transition ${}^4I_{15/2} \rightarrow {}^4I_{13/2}$ in the f shell are shown. The energy transfer is produced by Coulomb interaction. (3) Multiphonon nonradiative capture of the electron by the D^0 defect. The dashed curve represents the adiabatic potential in the intermediate (virtual) state of the DRAE transition (2).

the excess energy of the order of the Franck-Condon shift can be easily transmitted to local vibrations of the defect D.

The probability for the DRAE process can be expressed as[22]

$$w_A(R_0)=\frac{16\pi}{27}\frac{e^4 d_{f'f}^2}{\varepsilon_\infty^2(\kappa a)a^4 h^2\omega_p}I(kT)\exp\left(-\frac{2R_0}{a}\right). \quad (1)$$

In this expression the multiphonon factor $I(kT)$ is given by

$$I(kT)=(h\omega_p/4\pi SkT)^{1/2}\exp(-\varepsilon_{\rm ac}/kT). \quad (2)$$

$d_{f'}$ denotes dipole moments of transitions in the f shell of the erbium ion, κ^{-1} is the localization radius of the electron on a defect, a is the localization radius of a state in the conduction-band tail, ω_p is the local phonon frequency, R_0 is the distance between the electron in the band tail and the D center, and $\varepsilon_\infty$is the high-frequency dielectric constant.

The activation barrier for the DRAE process is (see Fig. 7)

$$\varepsilon_{\rm ac}=(\varepsilon-\Delta_{f'f}-Sh\omega_p)^2/4Sh\omega_p. \quad (3)$$

The Huang-Rhys factor S is determined by the difference of optical absorption and luminescence energies (Franck-Condon shift)

$$S=(\varepsilon_{\rm opt}-\varepsilon_l)/2h\omega_p. \quad (4)$$

The probability for the defect-related radiative transition (light emission) is given by

$$w_R(R_0)\approx\frac{128e^2\omega^3\exp(-2R_0/a)}{9\varepsilon_\infty c^3 h(a\kappa)^5\kappa^2}. \quad (5)$$

The ratio of the two rates is

$$\frac{w_A(R_0)}{w_R(R_0)}\approx\frac{\pi}{24}\frac{e^2\kappa}{\varepsilon_\infty h\omega}(\kappa\lambda)^3(d_{f'f}\kappa)^2 I(kT), \quad (6)$$

where the wavelength of erbium emission is

$$\lambda=\frac{hc}{E_{f'f}}. \quad (7)$$

The values $\varepsilon_{\rm opt}=1.05$eV, $\varepsilon_l=0.85$ eV, and $h\omega_p\approx50$ meV lead to a Huang-Rhys factor $S\approx2$ and $\varepsilon_{\rm ac}\approx6$ meV. Inserting these numbers in Eq. (6) shows that the DRAE process is more effective than the radiative electron capture:

$$\frac{w_A(R_0)}{w_R(R_0)}\approx27. \quad (8)$$

The competing nonradiative multiphonon transition (3) has an activation energy threshold ε_b determined by the energy between the bottom of the D^0+e potential and its crossing point with the D^- potential (Fig. 7):

$$\varepsilon_b=(\varepsilon-Sh\omega_p)^2/4Sh\omega_p, \quad (9)$$

which in our case is $\varepsilon_b\approx1.8$ eV. For such high activation energy multiphonon transitions should take place via thermally activated tunneling. Therefore, their probability is strongly temperature dependent and, as will be shown, it is this dependence that controls the thermal quenching of erbium luminescence.

Since the quantum yield of the intrinsic luminescence in a-Si:H(Er) is very low in our samples (~2%), the recombination via defects is equal to the total generation rate αI_p and is practically independent of the temperature if we ignore the weak temperature dependence of the absorption coefficient. However, the distribution of the recombination between different channels will be temperature dependent and will in fact determine the temperature dependence of the erbium luminescence. From the discussion above we conclude that while at low temperatures the DRAE channel dominates, the role of multiphonon transitions increases considerably at sufficiently high temperatures. The temperature quenching of the erbium luminescence is then controlled by the competition of the DRAE and multiphonon processes.

We have calculated the temperature dependence of the thermally activated multiphonon tunnel process using the parameters given above. The calculations were performed in the frame of the Huang-Rhys model of two displaced parabolas:[23]

$$w_{\rm mp}=w_{\rm mp}^0\exp(-\phi), \quad (10)$$

where

$$\phi=\left\{-\frac{\theta}{2}+\ln\frac{1+\sqrt{1+\xi^2}}{\xi}-\sqrt{1+\xi^2}+\xi\cosh\left(\frac{\theta}{2}\right)\right\}\frac{\varepsilon_T}{h\omega_p}, \quad (11)$$

$$\theta=\frac{h\omega_p}{kT},\qquad \xi=\frac{Sh\omega_p}{\varepsilon_T\sinh(\theta/2)}.$$

In this model the energy of thermal excitation is

$$\varepsilon_T=(\varepsilon_{\rm opt}+\varepsilon_l)/2. \quad (12)$$

Figure 5 compares the temperature dependence of the reciprocal capture probability $w_{\rm mp}^{-1}$ with the temperature dependence of the intensity of erbium luminescence. The absolute value of the probability was adjusted to the experimental curve at low temperatures, where the luminescence intensity is nearly temperature independent. From the similarity of both curves we conjecture that temperature quenching of the erbium luminescence at higher temperatures is controlled by the onset of multiphonon processes.

In this model of the erbium ion excitation it is also understood why the temperature dependence of the erbium emission and of the intrinsic emission of a-Si:H are different. At low excitation flux the intrinsic luminescence in a-Si:H is assumed to result from recombination via a tunneling process of geminate electron-hole pairs, which thermalize into the band tails close to the site where they were generated. This process is limited by diffusion of the faster carriers, usually electrons, which escape the characteristic interaction volume for such tunneling. The corresponding time is determined by $\tau_D\approx R^2/D_n$, where R is the interaction radius for radiative tunneling and D_n the diffusion coefficient. In amorphous silicon

$$D_n=D_{n0}\exp(-E_t/kT), \quad (13)$$

where E_t is the characteristic activation energy of electron localized in the conduction-band tail to the mobility edge in the range of 0.13–0.15 eV. The generation rate of photons that contribute to the intrinsic photoluminescence is

$$I_{\mathrm{eh}}=\frac{\alpha I_p \tau_D}{\tau_r}, \tag{14}$$

where τ_r is the radiative lifetime of the band-to-band transition. It is therefore the temperature dependence of the diffusion controlled by τ_D, which determines the quenching of the intrinsic luminescence.

In order to discuss the dependence of the erbium luminescence on the pump intensity we assume that in equilibrium the majority of dangling bonds are in the D^- state since the samples are n type. In this case illumination creates D^0 centers due to capture of holes and these D^0 centers capture electrons by radiative, DRAE, and multiphonon processes. Here we want to return to the experimentally found sublinear dependence of the emission intensity on the excitation intensity I_{ex} (Fig. 3). Slight deviations from the linear dependence may be observed when, besides the dominant linear recombination term, additional nonlinear recombination terms of higher order exist. In the case of a-Si:H such processes could be band-to-band recombination or band-to-band Auger processes. Since their contribution is expected to be rather small we will neglect them for the following. Assuming that practically all electron-hole pairs recombine via defects we arrive at coupled equations for the concentrations of D^0 defects (N_D^0) and erbium ions in the excited ${}^4I_{13/2}$ state (N^*):

$$\frac{dN_D^0}{dt}=\alpha I_p - w_R N_D^0 - w_{\mathrm{mp}} N_D^0 - w_A N_D^0\left(1-\frac{N^*}{N_0}\right), \tag{15a}$$

$$\frac{dN^*}{dt}=w_A N_D^0\left(1-\frac{N^*}{N_0}\right)-\frac{N^*}{\tau}, \tag{15b}$$

where α is the absorption coefficient, I_p the pump intensity, τ the lifetime of erbium ions in the excited state, and w_r, w_A, and w_{mp} the probabilities for radiative, DRAE, and multiphonon processes, respectively, averaged over R_0 [Eqs. (1), (6), and (10)]. The terms in brackets account for the saturation of erbium ions: the DRAE process is blocked if an erbium ion is already excited.

Steady-state conditions lead to an approximate expression for the luminescence intensity from the erbium ions with a radiative lifetime τ_R:

$$I_{\mathrm{PL}}=\frac{N^*}{\tau_R}=\frac{\dfrac{w_A}{w_R+w_{\mathrm{mp}}+w_A}\alpha I_p N_0}{\dfrac{w_A}{w_R+w_{\mathrm{mp}}+w_A}\alpha I_p+\dfrac{N_0}{\tau}}\,\frac{1}{\tau_R}, \tag{16}$$

which gives the exact results in the limiting cases of weak and strong excitation. In the linear regime under weak excitation Eq. (16) reduces to

$$I_{\mathrm{PL}}=\frac{w_A}{w_R+w_{\mathrm{mp}}+w_A}\,\alpha I_p\,\frac{\tau}{\tau_R}. \tag{17}$$

These equations are valid for excitation with photon energies below and above the band gap, though the absorption coefficients will be different.

As discussed before, the main contribution to the temperature dependence of I_{PL} comes from the factor $w_A/(w_R+w_{\mathrm{mp}}+w_{\mathrm{A}})$ due to the redistribution of recombination between multiphonon and DRAE channels, other factors are less temperature dependent. In the saturation regime under strong excitation we obtain

$$I_{\mathrm{PL}}=\frac{N_0}{\tau_R} \tag{18}$$

and the luminescence of the erbium ions depends weakly on temperature as pointed out earlier.[15]

For the emission due to radiative transitions into the defects (transitions 1 in Fig. 7) we derive

$$I_{\mathrm{PL}}^D=\frac{w_R}{w_R+w_{\mathrm{mp}}+w_A\left(1-\dfrac{N^*}{N_0}\right)}\,\alpha I_p\,. \tag{19}$$

At low excitation where $N^*\ll N_0$ the defect luminescence I_{PL}^D increase with the pumping power is much weaker than at high I_p because the DRAE process dominates below room temperature ($w_A\gg w_R, w_{\mathrm{mp}}$). This means that the defect luminescence should be weak for low pump rates but should increase after the erbium ions are saturated. Although the dependence of the defect luminescence on the pump intensity was not studied in this work within the linear regime our earlier results[21] confirm this conclusion.

For the discussion of the frequency-resolved measurements we change in Eqs. (15) dN_D^0/dt to $i\omega N_D^0$ and dN^*/dt to $i\omega N^*$. However, since capture of electrons by D^0 centers occurs significantly faster than the relaxation of excited erbium ions, we can neglect $i\omega N_D^0$ in Eq. (15a) and obtain for the erbium luminescence in the linear regime:

$$I_{\mathrm{PL}}=\frac{\dfrac{w_A}{w_R+w_{\mathrm{mp}}+w_A}\alpha I_p}{1+i\omega\tau}\,\frac{\tau}{\tau_R}. \tag{20}$$

The real part yields the in-phase luminescence intensity as

$$\mathrm{Re}(I_{\mathrm{PL}})=\frac{\dfrac{w_A}{w_R+w_{\mathrm{mp}}+w_A}\alpha I_p}{1+\omega^2\tau^2}\,\frac{\tau}{\tau_R}. \tag{21}$$

This result clearly demonstrates that the total lifetime of the erbium ion in the excited state is not directly connected to the probability of the DRAE transition and can be weakly temperature dependent at temperatures where the luminescence intensity changes strongly.

The intensity of the erbium luminescence follows the spectral dependence of the absorption coefficient as shown in Fig. 4. However, a plot I_{PL} versus α in Fig. 8 reveals additional features. Two regions of linear dependence of $I_{\mathrm{PL}}(\alpha)$ can be distinguished: in the region of defect absorption where only one electron is generated per absorbed photon and in the region of band-to-band absorption where electrons and holes are excited. The data point to a higher efficiency of erbium luminescence in the region of defect absorption.

The discussion so far was based on the assumption that all the generated carriers lead to generation of D^0 states which acquire their charge again by recombination [see Eq. (15a)].

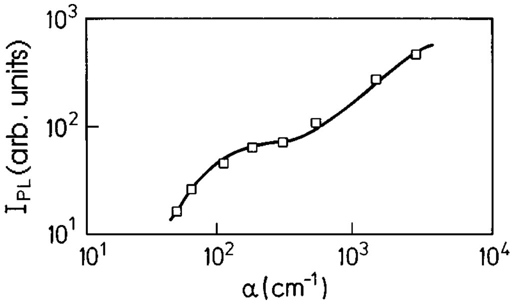

FIG. 8. Photoluminescence intensity as a function of absorption coefficient α (same data as in Fig. 4).

However, this assumption is correct only in the case of absorption of light by defects when only one type of free carrier, electrons, is generated. In the case of band-to-band absorption both electrons and holes are generated and only part of the recombination flux is described: namely, the capture of holes by the D^--states with subsequent capture of electrons. There exist other paths for recombination, like band-to-band recombination or nonradiative transitions involving D^+ states. We do not want to discuss these in detail, but it is obvious that in the case of interband absorption only part of the total generation rate αI_p leads to the formation of D^0 centers. This loss can be accounted for by modifying αI_p in Eqs. (15)–(21) by a factor η where $\eta=1$ for direct absorption and $\eta<1$ for band-to-band absorption. In agreement with the experiment we expect, therefore, a reduction of the erbium luminescence in the interband absorption region (Fig. 8).

V. CONCLUSIONS

a-Si:H(Er) exhibits pronounced photoluminescence at 1.54 μm. The primary step of the excitation mechanism is the absorption in the amorphous matrix and thus the creation of free carriers. We propose that the energy transfer from the free carriers to the Er ions occurs by Auger excitation associated with the capture of band-tail electrons by neutral defects (D^0). Our calculations show that the probability of this DRAE process is larger than that of radiative defect recombination by an order of magnitude. The DRAE excitation process is consistent with our experimental results. In particular the temperature quenching of the PL intensity above 200 K can be explained by the competition between the DRAE excitation process and nonradiative multiphonon recombination at the defects.

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