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Mechanism of light-induced reactivation of acceptors in *p*-type hydrogenated gallium arsenide

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The mechanism of light-induced reactivation (LIR) of shallow substitutional acceptors in high-purity *p*-type hydrogenated GaAs has been investigated. Low-temperature photoluminescence was used to determine the dependence of the rate and extent of this effect on photon energy, illumination intensity, sample temperature, and chemical identity of the passivated acceptor impurities. The efficiency of LIR at $T = 1.7$ K increases sharply for photon energies greater than a threshold value of $E_t \approx E_g - 7.5 \pm 0.5$ meV, where E_g is the band-gap energy of GaAs. This energy corresponds approximately to the onset of acceptor-bound exciton absorption in the material. For $h\nu > E_t$, the initial LIR rate depends on the square of the light intensity, indicating a bimolecular reaction involving the photogenerated carriers. For sufficiently large values of the product of the light intensity, and the illumination time, the LIR process saturates. Both the extent of the subthreshold effect for $h\nu < E_t$, and the saturation level that is attainable for $h\nu > E_t$ are independent of the photon energy, excitation power, and exposure time in the investigated ranges of these quantities. The LIR effect is practically athermal for very weakly neutralized acceptor species (e.g., Mg), but it is thermally assisted for Zn, Si, and Ge acceptors which form more stable complexes with hydrogen. From these results it is inferred, that the LIR of acceptors is electronically stimulated, possibly via a recombination-enhanced vibrational excitation of the acceptor-hydrogen complexes. A kinetic model of the LIR process, which accounts for the experimental results by assuming a reverse reaction of electronically stimulated relaxation of hydrogen toward an acceptor (light-induced passivation), is proposed.

I. INTRODUCTION

We have recently reported that certain shallow substitutional acceptors in high-purity *p*-type hydrogenated GaAs undergo a reversible and, essentially, athermal light-induced reactivation (LIR) at cryogenic temperatures.[1] The LIR process is strongly dependent on the chemical identity of the hydrogen-passivated acceptor species. An inverse correlation has been observed between the susceptibility to LIR and the binding energies of the different acceptor-hydrogen complexes (A-H) in GaAs.[2,3] The occurrence of LIR implies thermodynamic metastability of the hydrogenated GaAs (GaAs:H). Recently, Tavendale *et al.*[4] have confirmed instability of acceptor passivation in *p*-type GaAs:H under minority-carrier injection by illumination with above-band-gap light at room temperature. Since passivation of impurities and other defects via hydrogenation is considered a promising processing technique in fabrication of semiconductor devices,[5] an understanding of the LIR effect is important for the prediction of the long-term stability of devices based on this technology. Furthermore, the apparent similarity of the LIR phenomenon in GaAs to light-induced degradation in hydrogenated amorphous silicon (known as the Staebler-Wronski effect)[6] suggests that the understanding of the former process may assist in the ongoing effort to establish the mechanism of the latter, which takes place in the more complex amorphous material system. In this context the possibility of investigating hydrogen-related processes in GaAs under distinctly decoupled conditions of either pure electronic or optical stimulation, such as LIR at cryogenic temperatures, or pure thermal excitation [e.g., acceptor reactivation during high-temperature anneals at $T > 300\,^\circ$C (Refs. 7–9)], is particularly interesting.

In the present paper results of a systematic experimental investigation of the LIR mechanism are presented. The LIR effect was monitored as a function of photon energy and power density of the excitation light, as well as the sample temperature and the chemical identity of acceptor impurities using low-temperature photoluminescence (PL). The dependence of the rate and extent of LIR on these experimental parameters confirms our previous tentative proposal[1] that the effect is electronically stimulated, possibly via the recombination-enhanced defect reaction (REDR) mechanism.[10,11] An important aspect of LIR at $T = 1.7$ K is the fact that for large enough values of the product of the light intensity and the illumination time the reactivation process saturates at a level which is independent of the photon energy and light intensity, and is significantly lower than a complete recovery of the original acceptor concentration in the as-grown crystal. We discuss possible mechanisms that can cause the saturation effect and suggest that it is due to chemical equilibrium between LIR and the reverse process of light-induced passivation (LIP) of acceptors by hydrogen. A kinetic model, which agrees quantitatively with the experimental data and which supports the equilibrium concept, is presented.

II. EXPERIMENTAL PROCEDURE

We have previously demonstrated the LIR effect for a variety of high-purity GaAS layers grown by molecular-beam epitaxy (MBE) and metal-organic chemical-vapor deposition.[3] In this paper, for clarity of presentation, we concentrate mainly on the crystals that have exhibited the strongest susceptibility to LIR, Si-doped and nominally undoped GaAs epitaxial layers grown by MBE on (311)A-orientated, Cr-doped semi-insulating GaAs substrates.[1-3]

The hydrogenation was performed by an ~30-min exposure to a 13.56-MHz hydrogen plasma with a power density of 0.40 W/cm^2 in a Texas Instruments A-24-D parallel-plate reactor at a hydrogen pressure of 750 mTorr. The sample temperature was maintained at 250 °C. After the plasma was extinguished the samples were thermally quenched by a rapid transfer from the hot reactor to room temperature.

The as-grown, hydrogenated, and light-reactivated crystals were characterized with low-temperature photoluminescence. The samples were mounted strain-free in a Janis "Super Vari-Temp" variable-temperature cryostat. The sample temperature can be controlled over the range of 1.7–~270 K by immersing the samples either in superfluid ^{4}He or in heated He vapor. The PL measurements reported here were all performed at $T=1.7$ K. The photoexcitation of the samples during the PL measurements and for the stimulation of the LIR process was achieved with an unfocused beam of either the 5145-Å (2.409 eV) line from an Ar$^+$ laser, or the (1.5239±0.0001)-eV line from an Ar$^+$ laser-pumped Exciton LDS 821 dye. The latter light source allowed near-band-gap excitation, thus minimizing the uncertainty in the effective sample temperature, which could result from thermalization of hot electrons photogenerated with an excess kinetic energy of about 0.9 eV when the 5145-Å line from an Ar$^+$ laser is used. In addition, an intracavity, two-element birefringent filter in the dye laser allowed investigation of the photon-energy dependence of the LIR effect over the tuning range of about 1.42–1.55 eV, with a linewidth better than ~0.1 meV.

The illumination intensity on the samples was adjusted to the required level with neutral-density filters in the range of approximately 0.01–360 mW/cm^2. In this paper we refer to low-excitation conditions as those existing during the PL measurements; the corresponding excitation power densities, labeled P_L are less than ~4.0 mW/cm^2. The term "high-intensity illumination" and the symbol I are reserved for LIR stimulation, where light intensities (I) in the 4.0–360-mW/cm^2 range are used. The luminescence was dispersed in an Instruments SA 1-m double spectrometer and detected by a thermoelectrically cooled GaAs photomultiplier tube, using the photon-counting technique. Typical instrumental resolution was 0.1–0.2 meV for PL measurements of the broad conduction-band-to-neutral acceptor (e-A^0), and neutral donor-to-neutral acceptor (D^0-A^0) transitions. In one experiment, described in Sec. III and shown in Fig. 1, a different operating mode was used. The integrated intensity of the acceptor-bound exciton transitions (A^0,X) in the spectral range of approximately 1.5122–1.5126 eV was monitored real-time, by measuring the PL signal with wide spectrometer slits as a function of the LIR illumination time under $I=P_L\leq\sim4.0$-mW/cm^2 excitation conditions, rather than $I\gg P_L$ photopumping levels. Only these weak-excitation data were used for quantitative kinetic modeling of the LIR process. Under such illumination conditions, the sample heating due to light absorption can be neglected even at $T=1.7$ K. This assumption is based on the observations that both the vapor pressure of the ^{4}He liquid phase in the cryostat ($p\approx9.5$ Torr at $T=1.7$ K) and the PL peak linewidths and intensities (related to the lattice temperature via phonon scattering) remain unchanged over the analyzed excitation range of 0.35–4.0 mW/cm^2.

III. RESULTS

Light-induced reactivation of acceptors in GaAs:H can be detected using a variety of PL features, as discussed in detail in Refs. 1 and 3. Briefly, changes in absolute and relative PL intensities of the exciton and conduction-band-to-acceptor and donor-to-acceptor recombination spectra provide a clear indication of acceptor reactivation. For example, the $I(D^0\text{-}A^0)/I(e\text{-}A^0)$, $I(D^0,X)/I(A^0,X)$, and $[I(D^+,X)+I(D^0\text{-}h)]/I(A^0,X)$ intensity ratios can be correlated with the electrical compensation in the material,[1] whereas intensity ratios of the (D^0-A^0) or (e-A^0) peaks induced by different acceptor species permit assessment of the relative susceptibility of the corresponding A-H passivating complexes to LIR.[3] In subsequent sections these and other properties of the PL spectroscopy are employed to investigate the LIR process as a function of various experimental conditions. The following notation is used to label the dominant PL transitions: conduction-band-to-acceptor, (e-A^0); donor-to-acceptor, (D^0-A^0); free-exciton, FE; neutral donor and neutral acceptor-bound excitons, (D^0,X) and (A^0,X), respectively; mutually unresolved ionized donor-bound exciton, (D^+,X) and donor-to-valence band, (D^0-h).

A. LIR dependence on light intensity (I) of above-band-gap illumination

A direct example of the LIR kinetics is demonstrated in Fig. 1, which shows evolution of the integrated intensity of the $(A^0,X)_{n=1}$ transition at 10-sec intervals as a function of the illumination time t_{il}, at specified excitation power densities I of the 5145-Å line from an Ar$^+$ laser. All measurements were performed on the same sample. Each curve was taken after an overnight anneal at $T\approx60$ °C, which was found to restore, within experimental accuracy, the excitonic and impurity-related PL spectra to the original hydrogen-passivated state after exposure to LIR.[1] The intensity of the (A^0,X) luminescence from the freshly annealed sample ($t_{\text{il}}=0$) was used to normalize each curve.

The different curves in Fig. 1 exhibit common general trends. The (A^0,X) intensity increases approximately linearly with the illumination time at the initial stages of the LIR process, but for large enough values of the (It_{il})

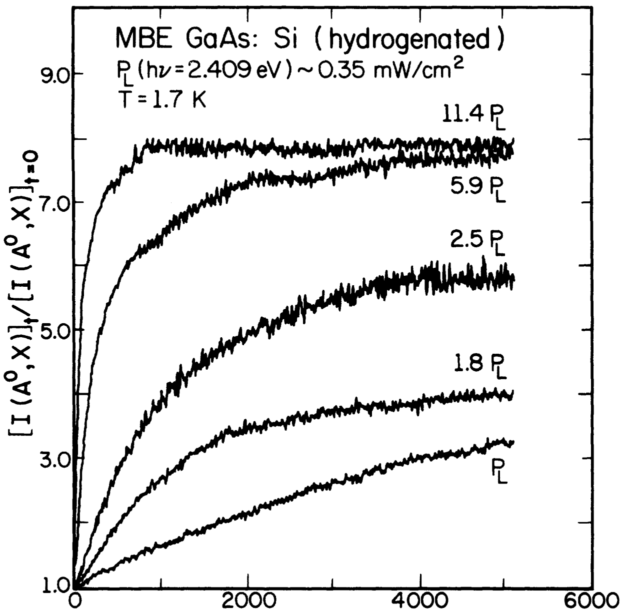

FIG. 1. Evolution of the (A^0,X) PL intensity with illumination time (t_{il}) at $T=1.7$ K for five different power densities (I) of the 5145-Å line from an Ar$^+$ laser. A strong dependence of the initial LIR rate on I, as well as saturation of the LIR process for sufficiently large (It_{il}) values, are evident.

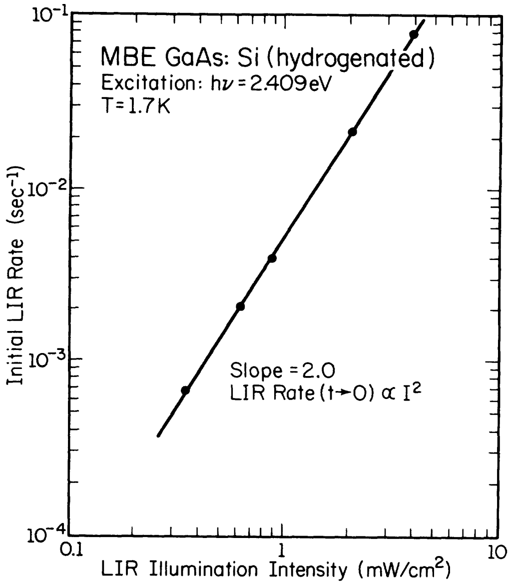

FIG. 2. Log-log plot of the initial LIR rate, given by $d(A^0,X)/dt$ for $t_{il}\rightarrow 0$ in Fig. 1, as a function of the illumination power density I of the 5145-Å line from an Ar$^+$ laser. The least-squares fit of the experimental points to a straight line gives slope of ~2.0, indicating that LIR is brought about by a bimolecular process of photopumped carriers.

product the (A^0,X) luminescence intensity saturates, as shown for the top curve in Fig. 1, recorded under the most intense light. The analysis of the entire PL spectrum after LIR indicates that the extent of LIR at $T=1.7$ K is significantly smaller than the complete recovery of the acceptor concentration in the as-grown crystal, which is attainable with thermal reactivation at high temperatures ($T>300\,^{\circ}$C).[7-9] We have pointed out previously that an appreciable contribution to the increase of the acceptor concentration during LIR at 1.7 K of the particular sample shown in Fig. 1 comes from the very weakly passivated Mg impurities, which were present only at a trace concentration before hydrogenation.[3]

After about 90 min of LIR by any of the low-intensity illumination processes shown in Fig. 1, the sample was exposed to a power density of ~360 mW/cm² of the 5145-Å laser light for an additional 30 min. No further changes in the entire PL spectrum or in the integrated intensity of the (A^0,X) transition, specifically, were observed for the sample saturated under the $I=11.4P_L \approx 4.0$-mW/cm² exposure (the top curve in Fig. 1). However, for all the other cases shown in Fig. 1 the additional high-power illumination caused both the (A^0,X) intensities and the overall PL spectra to resemble those of the saturated sample. Therefore it is inferred that for above-band-gap radiation the saturation level of the LIR process is independent of the illumination intensity. This result is indicated in Fig. 1 by convergence of the curve obtained under $I=5.9P_L$ illumination to the top, saturated curve for long t_{il}, and is more clearly seen for other curves on the log-log scale (not shown). This conclusion is further confirmed in another experiment described in Sec. III B below.

Saturation makes it difficult to observe the true kinetic characteristics of the LIR reaction. This complication has been avoided by determining the reactivation rates [given simply by the slopes of the (A^0,X) vs t_{il} curves in Fig. 1] as $t_{il}\rightarrow 0$ where, whatever the physical reason for saturation might be, its effects on the LIR process can be safely neglected. The dependence of the initial LIR rate on the excitation intensity is shown on the log-log scale in Fig. 2. The least-squares fit of the experimental points to a straight line gives a slope of 1.973. This implies that the initial rate of increase of the (A^0,X) luminescence grows superlinearly with the square of the illumination intensity $[d(A^0,X)/dt_{il} \propto I^2]$, indicating a bimolecular process of photopumped electrons and holes.

B. LIR dependence on excitation photon energy

A strong dependence of the reactivation rate on the light intensity was demonstrated in the preceding section. In order to determine whether the LIR effect indeed stems from creation of excess electron-hole pairs rather than from a photochemical reaction of direct photon absorption by A-H complexes or from phonons generated by thermalization of hot electrons photopumped by the 5145-Å laser light, we have examined the dependence of LIR on the incident photon energy $h\nu$. Photoluminescence spectra were measured at a fixed low-power excitation with the 1.5239-eV dye laser radiation. Between consecutive PL scans the sample was exposed for 5 min to a much more intense illumination at varying settings of the frequency tuning filter of the dye laser. Using any of the criteria for occurrence of LIR that are outlined above, the advancement of the reactivation process can be determined as a function of the photon energy.

A typical example of the experimental data for conduction-band-to-acceptor and donor-to-acceptor luminescence from the Si-doped epitaxial layer discussed so far, is presented in Fig. 3. The PL spectrum of the virgin hydrogenated sample (dashed line) changes only slightly after exposure to high-power excitation in the energy range of $1.422 \leq h\nu \leq 1.511$ eV. Independent of the excitation photon energy in this range or the illumination power density $I \approx 40-100$ mW/cm^2, the PL spectrum shown by the chained line in Fig. 3 is recorded. When the sample is subsequently illuminated with similar intensities (I) of higher energy light ($h\nu \geq 1.512$ eV) a drastic change occurs both in the detailed structure of the PL spectrum (solid line, Fig. 3) and in the absolute luminescence efficiency, which, in this case, approximately doubled. A similar variation of both the luminescence efficiency and the detailed PL pattern has been observed also in the excitonic spectral region and for other epitaxial layers.

Combined results of these experiments for several different samples are shown in Fig. 4. As mentioned above, weak reactivation was observed for excitation photon energies in the range of $1.442 \leq h\nu \leq 1.511$ eV. The extent of this change was independent of $h\nu$ and I for up to 2-h illumination of a given sample, but it varied from sample to sample within the range of approximately 10–30 % of the above-band-gap saturation value, as depicted in Fig. 4 by the horizontal dashed lines. At the photon energy $h\nu = E_g - 7.5 \pm 0.5$ meV, where the GaAs band-gap energy $E_g = 1.5192$ eV at $T = 1.7$ K is assumed,[12] there is a sharp transition to much more efficient LIR. This threshold energy E_t corresponds approximately to the energy of the (A^0,X) emission and, therefore, may be related to the onset of this bound-exciton absorption in GaAs.[12] The LIR saturation level reached with illumination energies $h\nu \geq E_t + 0.5$ meV has been found to be independent of both light intensity and photon energy up to $h\nu = 2.409$ eV, as shown in Fig. 4. This means that the isotropic lattice heating resulting from thermalization of electrons photoexcited with an excess energy as large as ~ 0.9 eV has no detectable effect on the reactivation process. The data in Fig. 4 also eliminate an electronic or vibrational excitation of an A-H complex via a direct photon absorption as a possible LIR mechanism. In those cases a resonant rather than step-function-like dependence of the reaction probability on the photon energy should be observed. Finally, if either the electron thermalization process or the direct photon absorption by A-H defects were responsible for LIR, a linear rather than the observed quadratic dependence on light intensity would be expected.

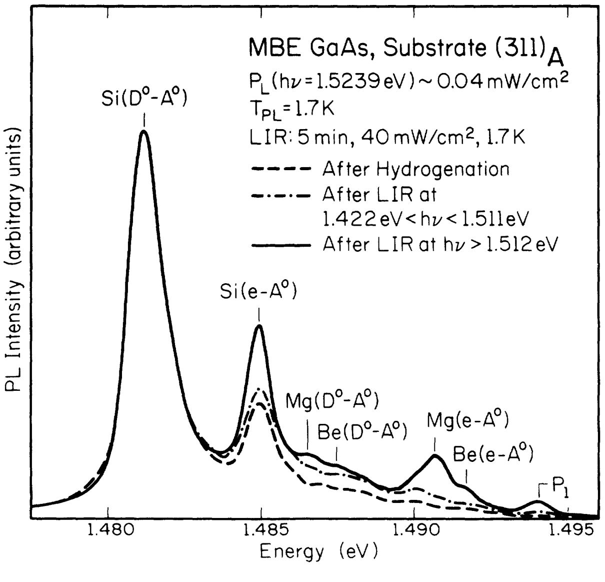

FIG. 3. Conduction-band-to-acceptor and donor-to-acceptor 1.7 K photoluminescence spectra of GaAs:Si crystal after hydrogenation (dashed line), after exposure to high-intensity light in the photon-energy range of $1.422 \leq h\nu \leq 1.511$ eV (dash-dotted line), and after a high-intensity, above-threshold illumination ($h\nu \geq 1.512$ eV). Increasing $I(\mathrm{Si}(e\text{-}A^0))/I(\mathrm{Si}(D^0\text{-}A^0))$ and $I(\mathrm{Mg}(e\text{-}A^0))/I(\mathrm{Si}(e\text{-}A^0))$ intensity ratios as well as the appearance of the P_1 transition allow monitoring of the LIR process.

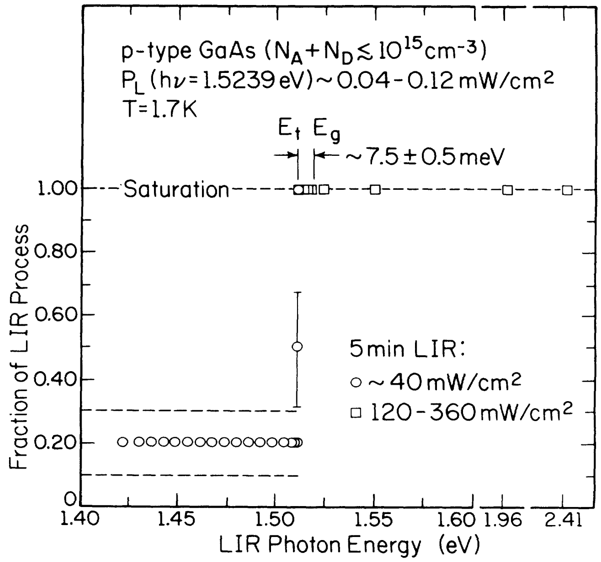

FIG. 4. Variation of the LIR efficiency at 1.7 K as a function of photon energy of the excitation source. A step-function-like dependence is apparent, with the threshold energy for LIR at 7.5±0.5 meV below-band-gap energy of GaAs. The dashed lines indicate the observed range of reaction advancement for different crystals exposed to below-threshold illumination. The limiting value reached within each energy range is independent of the photon energy in this range and the light intensity for up to 2-h exposure times investigated.

The results of this work demonstrate that the prerequisite for occurrence of efficient LIR is generation of excess electrons and holes in the material. The efficiency of this process is then determined by the product of electron and hole concentrations, giving rise to the observed dependence of the LIR rate on the square of photogeneration rate G, which for low excitation levels of the above-threshold radiation is proportional to light intensity (Fig. 2).

C. LIR dependence on sample temperature

We have reported previously that both the efficiency of hydrogen passivation and the susceptibility to LIR are strongly affected by the chemical identity of the passivated acceptor impurities.[2,3] This species dependence results from differences in the binding energies of various A-H complexes allowing determination of a relative passivation stability scale for shallow substitutional acceptors and acceptorlike defects in GaAs.[3] With increasing binding energy of an A-H complex the effectiveness of the acceptor neutralization increases, while the efficiency of the LIR process decreases. Isothermal studies of susceptibility to LIR for the specific MBE-grown GaAs:Si crystal discussed in earlier sections are summarized in Fig. 5 for the Mg and Si acceptors. The figure shows the evolution of the Mg(e-A^0) and Si(e-A^0) PL peak intensities, normalized to their respective initial values before LIR ($t_{il}=0$), as a function of the illumination time under the 4.0-mW/cm^2 illumination power density.[3] It is clear that both the initial rate and the extent of the LIR process, given roughly by the derivative as $t_{il} \rightarrow 0$ and the saturation value as $(t_{il} \rightarrow 0)$ of each curve, respectively, are significantly greater for Mg than for Si. From this and other cases it can be inferred that LIR of Mg is very efficient at $T = 1.7$ K compared to that of other acceptor species in GaAs.[3] It should be noticed that in the as-grown crystal the Mg acceptors are present only in a trace concentration relative to Si$_{As}$.[1,3] Furthermore, passivation of Si by H is much more extensive,[3] leading to $[\text{Si-H}]_0 \gg [\text{Mg-H}]_0$, where $[A\text{-H}]_0$ is the concentration of a given A-H complex in the freshly hydrogenated sample. From the greater extent of LIR for MG compared to Si, shown in Fig. 5, it is obvious that the LIR process does not reactivate all the available hydrogenated acceptor impurities and, therefore, it does not recover the original concentrations of different acceptor species, as asserted in Sec. III A. This important conclusion is further discussed in Sec. IV.

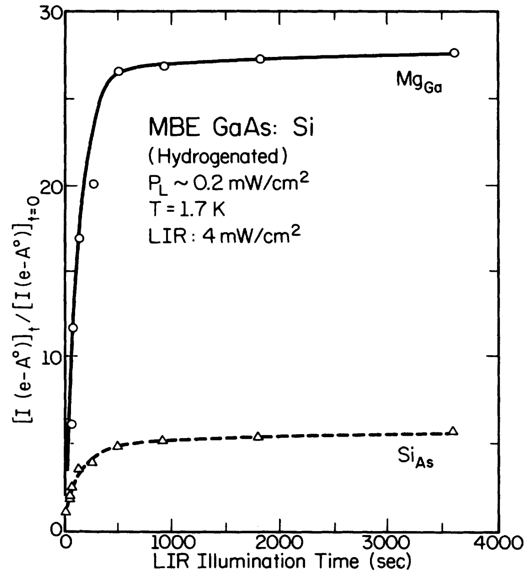

FIG. 5. Evolution of the (e-A^0) PL intensity with the above-band-gap illumination time during the LIR process of Mg and Si acceptors at $T_{\text{LIR}} = 1.7$ K. Both the rate and the saturation level of reactivation are greater for Mg than for Si acceptors, in spite of Mg being only a trace impurity in the as-grown crystal, and also very inefficiently hydrogenated relative to Si$_{As}$ acceptors. This result implies lower stability of the Mg-H passivating complex, and indicates that LIR does not lead to a complete recovery of the grown-in or passivated acceptor concentrations (Ref. 3)

In general, it is expected that with increasing sample temperature during LIR (T_{LIR}) the reactivation of the more stable A-H complexes will exhibit a stronger thermal activation. Thus, whereas LIR of MG should be only weakly, if at all, influenced by increasing T_{LIR}, reactivation of Be, Si, and Ge acceptors should be relatively enhanced at higher temperatures.[3] The photoluminescence technique is ideally suited to investigate these trends by monitoring relative intensities of acceptor-induced transitions due to all acceptor species simultaneously present in a given material. The effects of thermal contribution to the LIR process are demonstrated in Figs. 6 and 7 for two different samples. The data in Fig. 6 refer to the same GaAs:Si epitaxial layer as in Figs. 1–5. The variation of the PL spectrum of the as-grown crystal after hydrogenation and LIR at 1.7 K (the top three spectra in Fig. 6) has been analyzed extensively elsewhere,[1,3] although in the present work near-band-edge excitation at 1.5235 eV, instead of the 5145-Å Ar$^+$-laser line, has been used. Of interest here is the transformation of this spectrum with increasing T_{LIR} (the PL spectra were all measured under identical experimental conditions at $T_{\text{PL}} = 1.7$ K). As expected, the intensity ratios $I(\text{Mg}(e\text{-}A^0))/I(\text{Be}(e\text{-}A^0))$ and $I(\text{Mg}(e\text{-}A^0))/I(\text{Si}(e\text{-}A^0))$, which increased after LIR at 1.7 K diminish gradually at $T_{\text{LIR}} \approx 16$ and 100 K (the two bottom spectra in Fig. 6), due to a thermally assisted increasing reactivation efficiency of Be and Si relative to a very weakly thermally activated LIR of Mg acceptors.

An even more pronounced temperature effect on LIR of acceptors is shown in Fig. 7 for a nominally undoped GaAs crystal with residual C and Ge acceptor impurities. Again, the top three spectra correspond to the as-grown, freshly hydrogenated and the light-reactivated (at 1.7 K) samples. After hydrogenation the d_1–d_4(e-A^0) peaks induced by acceptorlike defect complexes characteristic of MBE-grown GaAs (Refs. 13 and 14) become quenched due to the extensive passivation of these centers.[3] In the 1.485–1.495-eV spectral range, where C(e-A^0) peak dominates in the as-grown crystal, new intense transitions have developed after hydrogenation as a result of a weak passivation of Mg and/or Be acceptors (the two cannot be mutually resolved in Fig. 7), and an enhanced contribution of the (D^0-A^0) emission relative to the (e-A^0) transitions.[1,3] Previously reported peaks labeled P_1 and P_2,[3,15] which are believed to originate in hydrogen-related acceptorlike shallow defects,[16] also emerge at the high-energy edge of the PL spectra in Fig. 7 after hydrogenation. The P_1 peak is detected in Fig. 6 as well. After LIR at 1.7 K an expected further enhancement of transitions due to weakly neutralized Mg and/or Be acceptors is observed, with the Mg and/or Be (D^0-A^0) and the

LIR-generated Mg and/or Be (e-A^0) peaks dominating the middle PL spectrum in Fig. 7. At higher sample temperature the LIR of Si, Ge, and, possibly, C acceptors is assisted more by the thermal contribution than that of the less stable Mg-H and/or Be-H passivating complexes. Consequently, the relative intensitites of the Mg and/or Be (D^0-A^0) and (e-A^0) peaks decrease dramatically, while Ge and C(e-A^0) transitions return to prominence, as shown in the two bottom spectra in Fig. 7 which were

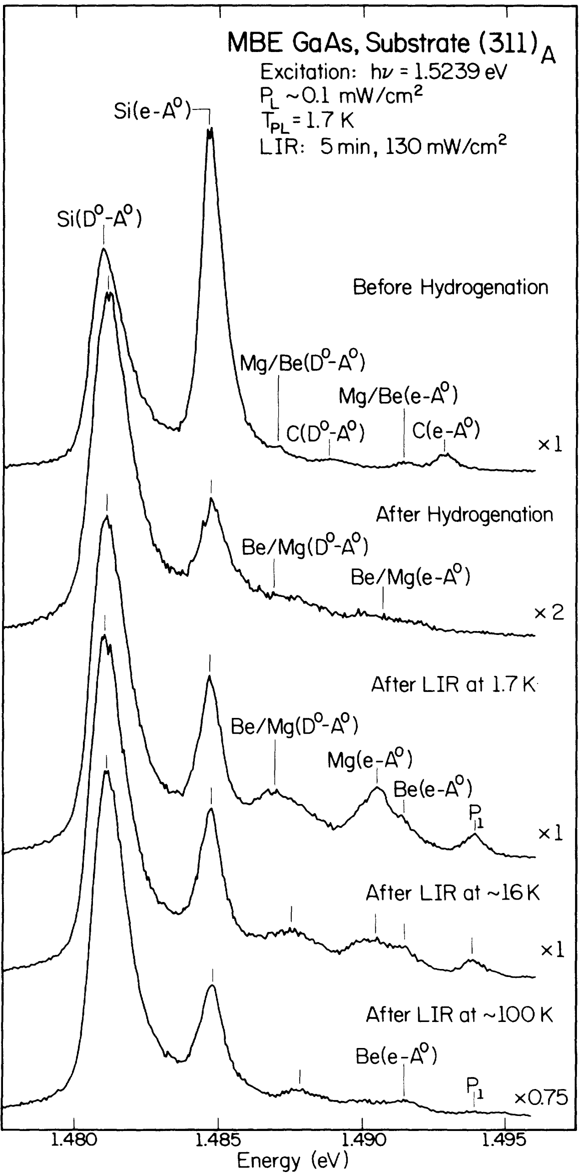

FIG. 6. Photoluminescence spectra measured at $T_{\mathrm{PL}}=1.7$ K of the as-grown (top spectrum) and hydrogenated GaAs:Si crystal. An inversion of the $I(\mathrm{Si}(D^0\text{-}A^0))/I(\mathrm{Si}(e\text{-}A^0))$ PL intensity ratio after hydrogenation is evident. After LIR at 1.7-K intensities of the peaks due to the weakly passivated Mg and Be acceptors increase dramatically (middle spectrum). At higher sample temperatures during LIR (bottom two spectra for LIR at ~ 16 and ~ 100 K) those transitions quench because of a more pronounced thermally assisted reactivation of the dominant Si acceptors.

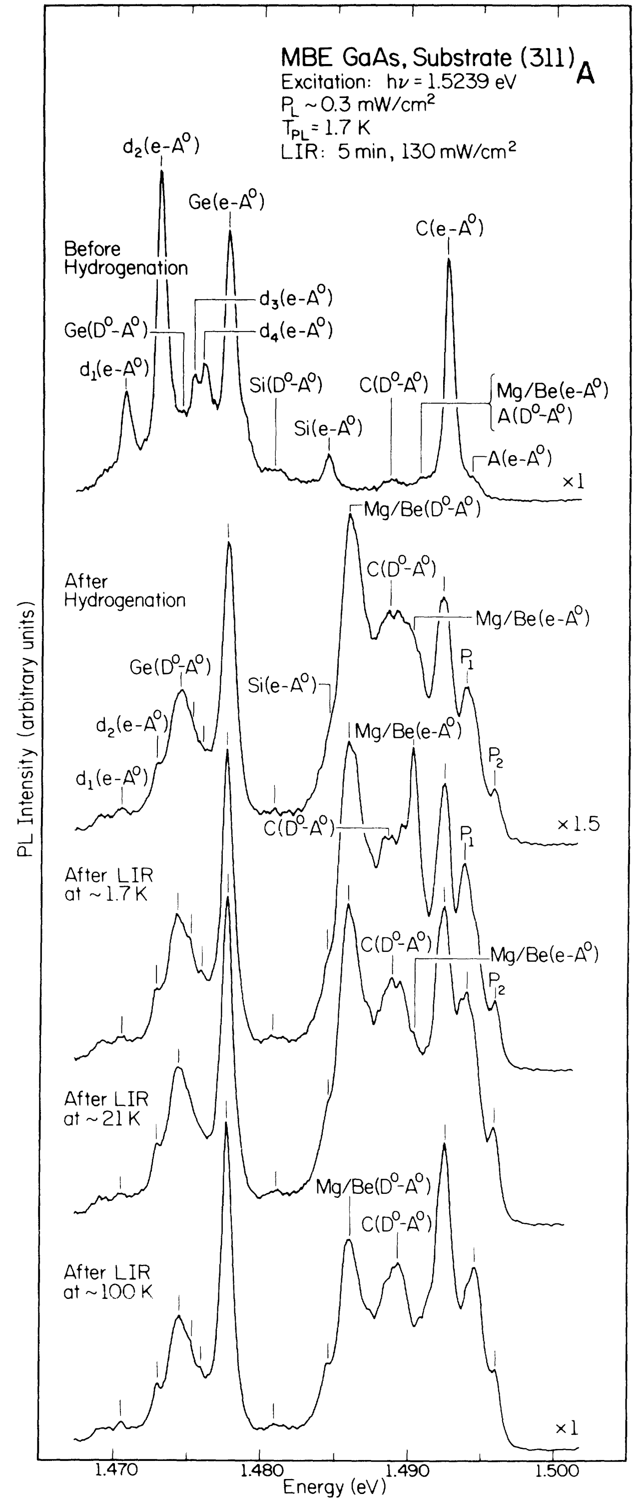

FIG. 7. Same as Fig. 6, but for a nominally undoped GaAs crystal grown by MBE on a $(311)_A$-oriented substrate. Carbon, Ge, and d_n defect complexes are the dominant acceptors before hydrogenation (top spectrum). Passivation of the d_n defects is very efficient relative to that of the substitutional acceptors present, and no LIR of these defects is observed up to $T_{\mathrm{LIR}} \approx 100$ K (bottom spectrum). Transitions in the 1.485–1.495-eV spectral region appear after hydrogenation and become further enhanced after LIR at 1.7 K, but they quench after LIR at $T_{\mathrm{LIR}} > 20$ K.

measured after LIR at ~21 and ~100 K. From Fig. 7 it can be inferred that no measurable LIR of the d_n acceptorlike defects takes place in the investigated temperature range, confirming their very stable passivation relative to that of the shallow substitutional acceptors.[3]

A general trend in all samples studied is that LIR at elevated temperatures causes a gradual quenching of those PL transitions which initially gained intensity after hydrogenation and the low-temperature LIR because of the inefficient passivation of the related impurities and defects. In contrast, transitions due to more extensively neutralized centers intensify at higher T_{LIR} because of a stronger thermal activation. Thus, with increasing T_{LIR} the PL intensity ratios between different acceptor-induced transitions tend toward those found in the as-grown material. This behavior is demonstrated clearly in Fig. 8 for the $I(\mathrm{Si}(e\text{-}A^0))/I(\mathrm{Mg}(e\text{-}A^0))$ intensity ratio of the hydrogenated GaAs:Si crystal. This ratio exceeded ~100 in the as-grown material. After hydrogenation, the ratio dropped to ~16 because of ineffective Mg passivation, and it decreased further to ~2.3 after LIR at 1.7 K due to extensive reactivation of weakly passivated Mg acceptors. However, the $I(\mathrm{Si}(e\text{-}A^0))/I(\mathrm{Mg}(e\text{-}A^0))$ intensity ratio increases gradually back to >20 with increasing sample temperature during LIR [the actual value is difficult to determine precisely, because the $\mathrm{Mg}(e\text{-}A^0)$ peak is barely detectable for the highest T_{LIR} studied].

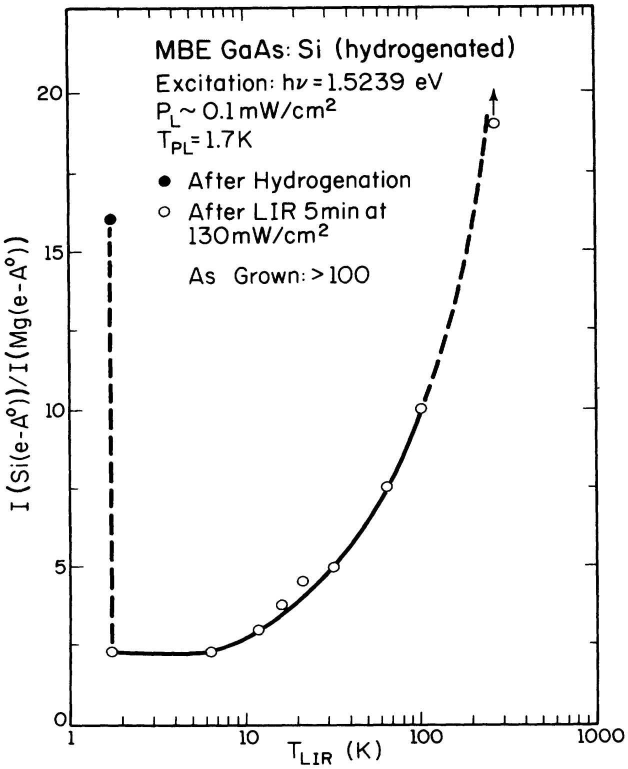

FIG. 8. Temperature dependence of the relative susceptibility to LIR of Mg and Si acceptors. Magnesium species are weakly passivated and thereby efficiently reactivated at $T=1.7$ K. At higher temperatures LIR of the Si acceptors becomes more efficient due to a stronger thermal activation.

The temperature dependence of LIR provides an additional important insight into the mechanism of reactivation. We have noticed that exposure to an above-threshold light at $T_{\mathrm{LIR}} > \sim 60$ K cause permanent changes in the passivation state of a material, as opposed to an almost completely reversible effect at lower T_{LIR}.[1] This observation may indicate that the thermal contribution not only assists in dissociation of more stable A-H complexes, but also, conceivably, enhances migration of hydrogen atoms in the crystal. Hydrogen diffusion facilitates formation of electrically and chemically inert, and relatively immobile H_2 molecules or hydrogen clusters, thus irreversibly removing H from the LIR repassivation reaction cycle, but not from the crystal.[17] When the data from Fig. 8 are plotted on a log vs $1/T$ scale, a sharp increase of the slope in the vicinity of $T_{\mathrm{LIR}} \approx 60$ K is clearly distinguished, as shown in Fig. 9. Since the slope of such an Arrhenius-type plot is, at least qualitatively, related to the activation energy of the reactivation process, a sudden change in the slope is consistent with the assumption that different reaction mechanisms are operative in the two temperature regimes.

The proposed scenario of temperature-assisted irreversible hydrogen recombination is further supported by data in Fig. 10, which shows two independent PL intensity ratios as a function of T_{LIR} for the same sample as in Fig. 9. Both quantities shown are, under our experimental conditions, directly related to the compensation ratio N_D/N_A in the crystal,[1,18] where N_D and N_A are concentrations of the electrically active donors and acceptors, respectively. In the as-grown crystal $N_D/N_A \approx 0.18$,[19] and it results in PL intensity

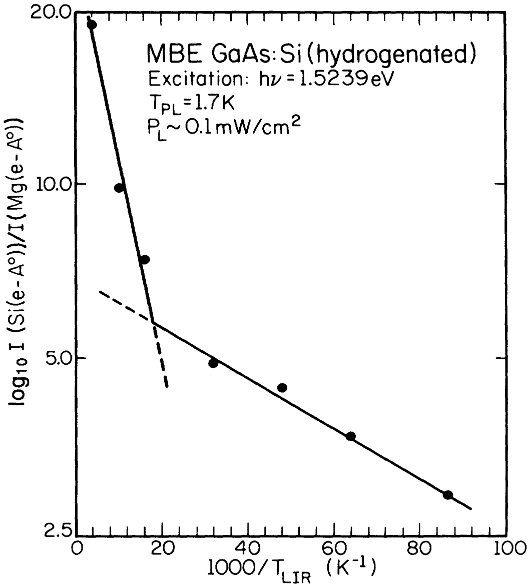

FIG. 9. Data from Fig. 8 for $T_{\mathrm{LIR}} > 10$ K are plotted on a log vs $1000/T$ scale. An abrupt increase of the slope in the vicinity of $T_{\mathrm{LIR}} \approx 60$ K may indicate a change in activation energy of the reactivation process, implying that different mechanisms are operative in each temperature regime. Light-induced reactivation at $T_{\mathrm{LIR}} > 60$ K causes irreversible modifications of the PL spectra, possibly due to the $\mathrm{H}+\mathrm{H} \rightarrow E\text{-}\mathrm{H}_2$ reaction.

ratios $I(\mathrm{Si}(D^0\text{-}A^0))/I(\mathrm{Si}(e\text{-}A^0))\approx0.7$ and $[I(D^+,X)+I(D^0\text{-}h)]/I(A^0,X)\approx0.05$. Both intensity ratios in Fig. 10 increase after hydrogenation due to passivation of acceptors, which increases the compensation ratio.[1] In fact, under our plasma conditions neutralization of acceptors in p-type materials is so extensive that compensation renders the hydrogenated samples too resistive for reliable variable-temperature Hall-effect measurements.[9] After LIR at 1.7 K the compensation is reduced slightly, but it gradually increases at higher LIR temperatures in the range of $1.7<T_{\mathrm{LIR}}M\sim60$ K. For $T_{\mathrm{LIR}}>\sim60$ K both intensity ratios in Fig. 10 decrease abruptly, and for reactivation at $T>300\,°\mathrm{C}$ the PL spectrum of the as-grown crystal is recovered. These results can be consistently explained assuming that H acts as a compensating donor in p-type crystals thus increasing the N_D/N_A ratio as long as [H] concentration increases due to LIR namely for $T_{\mathrm{LIR}}<60$ K. However, at higher temperatures the hydrogen recombination reaction $\mathrm{H}+\mathrm{H}\rightarrow\mathrm{H}_2$ reduces [H] by forming electrically inactive H_2 molecules, while LIR continues to generate electrically active acceptors. Accordingly, the N_D/N_A ratio decreases toward its original low value, causing a corresponding drop in the related PL intensity ratios, as shown in Fig. 10.

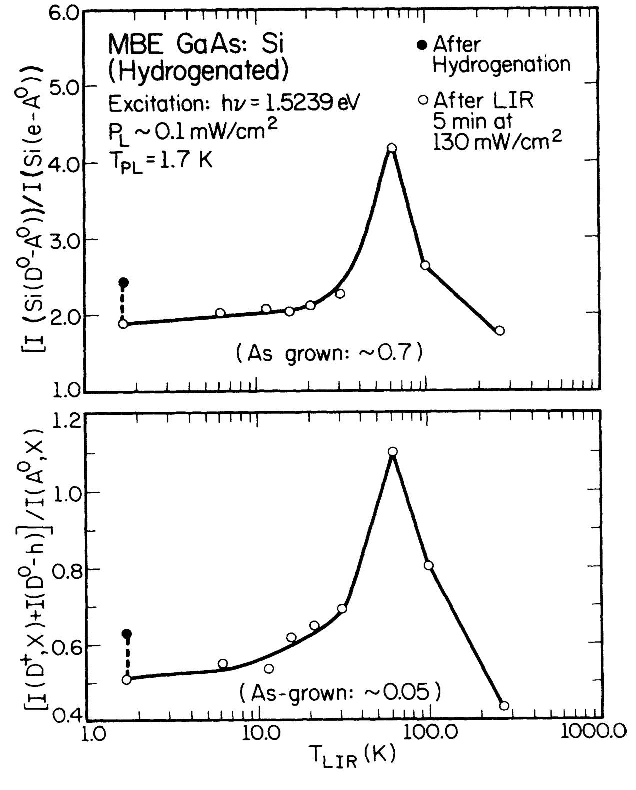

FIG. 10. Two independent PL intensity ratios, which are directly related to the compensation ratio N_D/N_A in the material, are shown as a function of the sample temperature during LIR. An initial increase of both ratios with temperature for $T_{\mathrm{LIR}}<\sim60$ K is explained by increasing N_D/N_A due to compensating action of the reactivated H donors. The subsequent decrease of compensation at $T_{\mathrm{LIR}}>60$ K may result from thermally assisted migration of H and its recombination to electrically inactive H_2 molecules.

IV. DISCUSSION

Any discussion of the LIR mechanism requires prior knowledge of the correlation between the measured PL intensities and the actual concentrations of the optically active acceptor impurities, so that quantitative information can be derived from Figs. 1 and 2 to model the reactivation kinetics. The rate equations of the FE and (A^0,X) concentrations are given by

$$dN_{FE}/dt=g-N_{FE}(W_{FE}+C_AN_A+W_0) \tag{1}$$

and

$$dN_{AX}/dt=C_AN_{FE}N_A-W_{AX}N_{AX}\ , \tag{2}$$

where N_{FE}, N_{AX}, and N_A are the concentrations of the FE, (A^0,X), and neutral acceptors, respectively; g is the free-exciton generation rate related to the excitation intensity, W_{FE} and W_{AX} are the radiative recombination rates of FE and (A^0,X), respectively, C_A is the FE capture rate at neutral acceptors, and W_0 represents all other competing radiative and nonradiative channels of FE annihilation and capture. Thermal dissociation of (A^0,X) complexes at 1.7 K is neglected in Eq. (2) because the localization energy of excitons on $(A^{\mathrm{II}})^0_{\mathrm{Ga}}$ and $(A^{\mathrm{IV}})^0_{\mathrm{As}}$ acceptors in GaAs is about 2.7 meV.[20] Assuming that the excess carrier thermalization and the FE formation, recombination, and capture by impurities and defects are much faster than LIR rate, momentary steady-state FE and (A^0,X) populations are maintained during LIR. Thus the integrated intensity of the (A^0,X) transition is

$$I(A^0,X)\propto N_{AX}=\frac{gC_A}{W_{AX}}\frac{N_A}{W_{FE}+C_AN_A+W_0}\ . \tag{3}$$

If we assume that the extent of LIR at 1.7 K is small compared to concentrations of the already active impurities and defects in a material, then all the parameters on the right-hand side of Eq. (3), except for the increasing N_A, remain roughly constant during reactivation. Furthermore, considering a low N_A in the passivated material, the multitude of the detected PL transitions, as well as many other radiative and nonradiative recombination processes conceivably taking place in the crystal $(W_{FE}+W_0)$ should be of the order of magnitude or greater than C_AN_A throughout the LIR process. Both assumptions should be very accurate in particular for $t\rightarrow0$, but still reasonable for greater t_{il} provided that the extent of LIR is small, as asserted above. It follows that the increase in absolute integrated intensity of the (A^0,X) luminescence during LIR (Fig. 1) is, under our experimental conditions, approximately proportional to the concentration of the reactivated shallow acceptor impurities. The data of Skromme *et al.*[14] and Lu *et al.*,[21] obtained under less controlled experimental conditions, namely for different samples and over a much larger concentration range, support the relaxation $I(A^0,X)\propto N_A$.

The characteristic features of the LIR effect, namely the existence of the near-band-gap photon energy threshold, photon-energy-independent saturation level, and the I^2 dependence of the initial reactivation rate, provide strong support for electronic stimulation of the LIR process. These observations, as well as a very low (if any) effective activation energy for LIR of certain acceptors (e.g., Mg, Be) at 1.7 K can be consistently interpreted in terms of recombination-enhanced kinetics.[10,11] In our case, because of the I^2 dependence, this mechanism involves a local vibrational excitation of the A-H complex via multiphonon recombination of *both an electron and a hole* virtually simultaneously at the same hydrogen-passivated defect center.[22] Based on the results from Figs. 1 and 2, and neglecting at this point the saturation effect, the rate equation for the initial LIR process can be written as

$$d\Delta N_A/dt = K_{\mathrm{LIR}} R N_{AH}(t) = k_{\mathrm{LIR}} R[N_{AH}(0)-\Delta N_A(t)]\ , \tag{4}$$

where $\Delta N_A(t)=N_A(t)-N_A(t=0)$ is the concentration of reactivated acceptors, $N_{AH}(0)$ and $N_{AH}(t)$ are, respectively, the initial (total) and momentary concentrations of the A-H complexes that are susceptible to LIR, k_{LIR} is the LIR rate constant, and $R \propto I^2$ is the recombination rate of electron-hole pairs at an A-H defect complex. The rate constant for such a recombination-enhanced process is given by[10,11]

$$k_{\mathrm{LIR}} \propto h \exp[-(E_{T-E_R})/kT]\ , \tag{5}$$

where h is the probability that the recombination energy, E_R is routed into the dissociative vibrational mode of the A-H complex, and E_T is the activation energy for pure thermal reactivation of the acceptor. Only a single value of E_T($\sim$1.6 eV for Zn acceptors) has been reported so far.[7,8] The apparent activation energy for LIR is reduced by $E_R \le F_n - F_p$, where F_n and F_p are the quasi-Fermi levels for electrons and holes, respectively. In GaAs, under a low excitation at 1.7 K, $E_R \le E_g \approx 1.52$ eV, thus explaining the high efficiency of LIR for Mg acceptors, which are known to have a lower affinity to H than any other substitutional acceptors.[3] Light-induced reactivation of passivated Zn acceptors has not been observed at 1.7 K, but they are slightly reactivated at $T \approx 20$ K.[3] Similarly, the dependence of E_T on the chemical identity of the acceptor species accounts for the variation of PL spectra with increasing LIR temperature, as discussed in Sec. III C and demonstrated in Figs. 6–8. The concentration of the energy $E_R \approx E_g$ into a specific vibrational mode along the dissociation reaction coordinate of the A-H complex requires that the capture of both an electron and a hole occurs within a single lifetime of the recombination center Δt before the energy transferred upon a single-carrier capture event dissipates among other, nonreactive vibrational modes.[10,11,22] In principle, simultaneous trapping of an electron-hole pair as a defect-bound exciton is also possible at cryogenic temperatures. However, probability for this event is rather low under very weak excitation conditions, such as those employed in the experiment shown in Fig. 1.

It should be emphasized that as a precondition for significant recombination enhancement, the A-H complex should form a deep state in the energy gap, so that its recombination lifetime is long and there is a strong electron-phonon coupling.[10,11] To our understanding, this requirement is inconsistent with the widely accepted Pankove model of acceptor passivation,[23] which implies a removal of impurity states from the forbidden gap upon hydrogenation via rearrangement of the defect structure and formation of stable bonds.[17] Furthermore, so far there is no direct spectroscopic evidence for a hydrogen-related deep center in GaAs PL which could put our proposal of the REDR mechanism on a firmer basis. Thus although the REDR model is consistent with all the available data related to the LIR effect, it should be qualified that other electronically stimulated mechanisms could be conceived as well.

A full kinetic model of LIR should account also for the effect of saturation. In principle, the following mechanisms may be responsible for saturation. First, in the framework of the REDR mechanism, the lifetime of photogenerated carriers and, therefore, their steady-state concentrations decrease upon reactivation of new acceptor and hydrogen trapping centers (we note that two traps, an ionized acceptor and a proton, are created for each reactivated A-H recombination center). Moreover, since the lifetime determines the diffusion length of carriers from the vicinity of the crystal surface, where the above-band-gap light is absorbed, into the bulk of an epitaxial layer where LIR presumably occurs,[1] the reaction volume effectively shrinks due to the shunting effect of the reactivated defects. This self-limiting behavior of LIR is qualitatively analogous to that proposed by Stutzman, Jackson, and Tsai[24] to explain saturation of the Staebler-Wronski effect in *a*-Si:H.[6] Second, the LIR process may be completed when all the susceptible A-H complexes become reactivated $[\Delta N_A(t\rightarrow\infty)\approx N_{AH}(0)]$. Finally, the saturation of LIR may result from thermodynamic equilibrium between the LIR and the reverse reaction of relaxation to the passivated state

$$(A\text{-H})^0 \rightleftarrows (A)^- + \mathrm{H}^+\ . \tag{6}$$

The fact that the saturation level is independent of both light intensity and photon energy (the latter affects the light penetration depth and, thus, the reaction volume) suggests that the first mechanism, although conceivably operative, is not dominant, especially when the extent of LIR is small relative to concentrations of other existing recombination centers. The observation that the saturation level does not vary with illumination intensity implies either (i) termination of the LIR process by depletion of $N_{AH}(0)$ or (ii) dependence of the relaxation reaction on carrier recombination rate R similar to that of the LIR effect [Eq. (4)]. The second requirement is intuitively consistent with the argument that, since the relaxation process presumably involves H diffusion toward an acceptor, its occurrence at 1.7 K indeed suggests a recombination enhancement. The great dissimilarity between the PL spectra of the as-grown and light-reactivated materials, the strong dependence of the latter on the LIR temperature, and the observation that the saturation level of LIR for different acceptor species depends on the binding energy of the A-H complexes rather than on the grown-in or passivated concentrations of the corresponding impurities (Fig. 5), precluding the LIR completion mechanism. Thus we propose existence of a LIP effect, which leads to the overall kinetic equation

$$d\Delta N_A/dt = k_{\mathrm{LIR}} R[N_{AH}(0) - \Delta N_A(t)] - k_{\mathrm{LIP}} R\, \Delta N_A(t) \ . \tag{7}$$

Equation (7) can be simplified to

$$d\Delta N_A/dt = R(k_{\mathrm{LIR}} + k_{\mathrm{LIP}})[\Delta N_S - \Delta N_A(t)] \ , \tag{8}$$

where $\Delta N_S \int k_{\mathrm{LIR}} N_{AH}(0)/(k_{\mathrm{LIR}} + k_{\mathrm{LIP}})$ is the equilibrium concentration of light-reactivated acceptors at saturation.

From Fig. 2 we have determined that the LIR effect involves a bimolecular process of recombination of both an electron and a hole at the A-H defect sites, described by

$$R = Bnp \ , \tag{9}$$

where n and p are steady-state concentrations of electrons and holes, respectively, and B is a recombination coefficient. In the temperature regime of carrier freezeout the condition $n \approx p$ is maintained even under low-level excitation, namely for $n,p \ll N_R(t_{\mathrm{il}})$, where $N_R(t_{\mathrm{il}})$ is the momentary concentration of all available recombination centers. Thus, at cryogenic temperatures

$$n \approx p = G t_{e,h} = I t_{e,h}/(Lh\nu) \ , \tag{10}$$

is maintained, where G is the photogeneration rate, $t_{e,h} = [N_R(t) b_{e,h}]^{-1}$ is the electron/hole lifetime, b_e and b_h are the capture rate coefficients for electrons and holes, respectively, $h\nu$ is the photon energy, and L is the ambipolar diffusion length. Combining Eqs. (9) and (10) gives

$$R = \frac{B}{\beta_e \beta_h (Lh\nu)^2} \left[\frac{I}{N_R(t)} \right]^2 . \tag{11}$$

Mass balance of Eq. (6) requires $N_R(t) = N_R(0) + \Delta N_A(t)$, and Eq. (8) becomes

$$\frac{d\Delta N_A}{dt} = A I^2 \frac{\Delta N_S - \Delta N_A(t)}{[N_R(0) + \Delta N_A(t)]^2} \ , \tag{12}$$

where $A \int B/[b_e b_h (Lh\nu)^2]$. Integration of Eq. (12) yields the time dependence of ΔN_A

$$-\frac{\Delta N_A^2(t)}{2} - [2N_R(0) + \Delta N_S]\Delta N_A(t) + \{N_R^2(0) + \Delta N_S[2N_R(0) + \Delta N_S]\} \times \ln \left[\frac{\Delta N_S}{|\Delta N_S - \Delta N_A(t)|} \right] = A I^2 t_{\mathrm{il}} \ . \tag{13}$$

The kinetic model in Eq. (13) can be examined by defining a normalized dimensionless quantity

$$\Delta N_A(t) = \{[I((A^0,X))]_t - [I((A^0,X))]_{t=0}\}/[I((A^0,X))]_{t=0} \ . \tag{14}$$

Using experimental data from Fig. 1, we plot in Fig. 11 $\Delta N_A(t)$ vs t_{il} for five different illumination intensities, and compare these data with the kinetic curves predicted by the model (solid lines in Fig. 11). Good agreement is obtained over the entire illumination time range for four of the shown cases by adjusting the light intensity for each curve to the known value indicated in Figs. 1 and 11, while keeping the parameter A constant and allowing a small (±3%) variation in the values of ΔN_S and $N_R(0)$. A discrepancy between the experimental data and the model curve above $t_{\mathrm{il}} > 2000$ sec for $I = 1.8P_L$ in Fig. 11 is probably due to a sudden decrease in the laser power during this particular measurement. The parameter A is a kinetic constant of the LIR and/or LIP process and, therefore, should be identical for all curves. On the other hand, small fluctuations in ΔN_S and $N_R(0)$ for different scans are justified by experimental inaccuracies resulting from the assumption of complete reversibility of a material to exactly the same passivated state after each LIR cycle, and from the normalization procedure which depends critically on measurement uncertainty of the $[I(A^0,X)]_{t\to 0}$ PL intensity (estimated as about 5%). Best fits of Eq. (13) to the experimental data are obtained for $A = 4.49 \times 10^4$ cm^4/W^2 sec and $N_R(0) \approx \Delta N_S = 6.65 \pm 0.20$.

The nondispersive rate equation of the Staebler-Wronski degradation in a-Si:H, proposed recently by Bube and Redfield[25] (Eq. 9 in Ref. 25) as an extension of the original degradation model of Stutzmann, Jackson, and Tsai,[24] is a special case of our model [Eqs. (12) and (13)] when $\Delta N_A(t_{\mathrm{il}}) \gg N_R(0)$, namely when the carrier lifetime is dominantly controlled by recombination at the

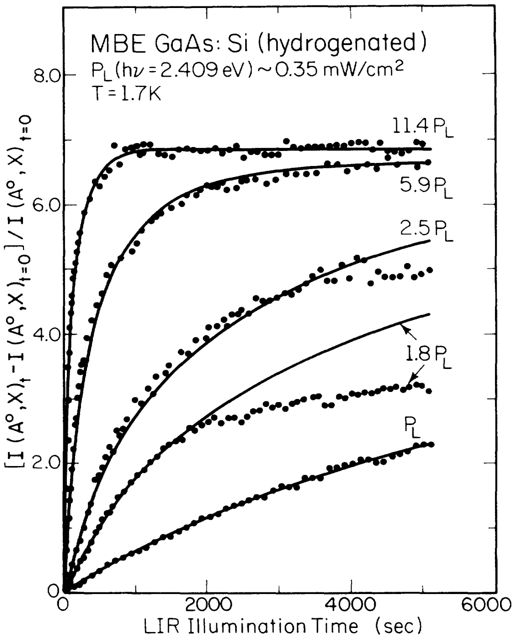

FIG. 11. Comparison of the experimental data for ΔN_A vs t_{il} (dots) with the kinetic model of LIR in Eq. (14) (lines). Good agreement is obtained using the indicated illumination intensities as the externally adjustable parameters for different curves. The other model parameters in Eq. (14) were constrained to $A = 4.49 \times 10^4$ cm^4/W^2 sec and $N_R(0) \approx \Delta N_S = 6.65 \pm 0.20$.

light-generated metastable defects. The result $N_R(0) \approx \Delta N_S$ obtained in this work for LIR in high-purity GaAs:H is consistent with our basic assumption that $\Delta N_A(t_{\mathrm{il}})$ is smaller than or, at most, comparable to the total concentration of all other, radiative and nonradiative centers [see discussion following Eq. (3) above]. Our model is general in the sense that it can be also applied without modifications to purely thermal processes of reactivation, degradation, or annealing where the carrier lifetime is immaterial and can be assumed constant $[\Delta N_A(t_{\mathrm{il}}) \ll N_R(0)]$, so that conventional first-order kinetics prevail.

V. CONCLUSIONS

A systematic study of light-induced reactivation of shallow substitutional acceptors in high-purity *p*-type hydrogenated GaAs has been reported. Dependence of this effect on photon energy, light intensity, crystal temperature, and the chemical identity of passivated acceptor species indicates that LIR is electronically stimulated. We have proposed one possible mechanism (REDR), in which LIR proceeds via carrier recombination-enhanced vibrational excitation of acceptor-hydrogen passivating complexes. Specifically, the process should involve simultaneous capture of both an electron and a hole at the same *A*-H defect site, giving rise to a superlinear dependence of LIR rate on the square of light intensity. Efficiency of LIR at $T = 1.7$ K is a step function of photon energy, with a sharp transition from a weak to a strong recombination enhancement at the photon threshold energy $h\nu = E_g - 7.5 \pm 0.5$ meV. This energy corresponds approximately to the onset of the (A^0, X) absorption in GaAs.

For some acceptor species (e.g., Mg, Be) the LIR process is very efficient at 1.7 K and it is not affected significantly by increasing the sample temperature during LIR. In the framework of the REDR mechanism this nearly athermal behavior indicates that the dissociation energy of the corresponding *A*-H complexes is approximately equal to E_g of GaAs at $T \sim 1.7$ K. For other acceptor species, such as Zn, Si, and Ge, efficiency of LIR increases with increasing temperature, indicating a small but finite species-dependent activation energy for dissociation of the corresponding *A*-H complexes. These trends comply with quantitative predictions of the recombination-enhanced dissociation kinetics.[10,11]

The LIR effect is reversible at $T_{\mathrm{LIR}} < \sim 60$ K, but permanent changes in a material take place under exposure to an above-threshold radiation at $T_{\mathrm{LIR}} > 60$ K. This behavior is accompanied by both an abrupt increase of the activation energy for LIR and a decrease in the compensation ratio of a material at $T_{\mathrm{LIR}} > 60$ K. We have explained these observations by proposing that an increasing T_{LIR} facilitates migration of reactivated H atoms (which act as compensating donors in *p*-type GaAs), eventually causing a reduction in concentration of the electrically active hydrogen in the crystal via formation of inert H_2 molecules.

Finally, we have discussed kinetics of the LIR reaction and, in particular, possible mechanisms for photon energy and light-intensity-independent saturation effect of LIR. We have proposed that saturation results from both a self-limiting process of carrier lifetime decay due to generation of light-reactivated acceptor and H recombination centers, and a chemical equilibrium established between the forward LIR process and a reverse reaction of light-induced passivation of acceptors. A quantitative kinetic model has been presented and found to agree well with the experimental data. This kinetic model offers a generalized approach to conventional and electronically stimulated first-order reactions and should be applicable to studies of analogous effects in other material systems, such as amorphous semiconductors.

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