

## Role of the hydrogen atom on metastable defects in GaAs

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Several deep-level defects in hydrogen-passivated GaAs doped with Si have been investigated. The defect transformation by atomic hydrogen confirms the assignment of these defects to metastable defects associated with a hydrogen atom. Thermal-annealing experiments under biased and unbiased conditions confirm that during hydrogenation a deep level at 0.60 eV below the conduction band is generated, as a metastable defect for the native deep level at 0.42 eV below the conduction band, and the complete passivation of the 0.42- or the 0.33-eV trap during hydrogenation is due to passivation of the trap by a hydrogen-atom-forming hydrogen-defect complex. The first-order kinetics permits a precise estimate of the formation and annealing frequencies  $\nu_f$  and  $\nu_a$  of the hydrogen-defect pair. The temperature-dependent values of  $\nu_a$  for the 0.60-eV trap satisfy the relation  $\nu_a = (0.82 \times 10^{13}) \exp[(-1.61 \pm 0.04 \text{ eV})/kT] \text{ s}^{-1}$ . We propose that this activation energy could be the value required for the release of a hydrogen atom bound to a point defect in GaAs.

### I. INTRODUCTION

It has been known that the exposure to hydrogen plasma results in electrical passivation of impurity states<sup>1-6</sup> and deep levels<sup>1,7,8</sup> as well as unreconstructed surface dangling bonds<sup>9</sup> in GaAs. The mechanism for the hydrogen passivation and the structures of the complexes that might form during hydrogenation have been studied theoretically and experimentally. Atomic hydrogen is found in a variety of configurations in GaAs. As an example, a suggestion<sup>1</sup> has been advanced that in *n*-type GaAs its charge state is neutral, while in *p*-type GaAs it may be ionized. Also, atomic hydrogen can interact with existing native defects during the diffusion, forming new defects.<sup>10</sup> For this reason, the use of atomic-hydrogen processing may ultimately prove to be more valuable in defect identification than the practical aspect of defect passivation. Compared with the great amount of knowledge on the role of atomic hydrogen in passivating the electrical activity of shallow donors and acceptors in GaAs, the microscopic identification of hydrogen-defect association that results in passivation has not been established for deep-level defects.

An interesting class of semiconductor-defect reactions involves metastable defect configurations such as the *DX* center in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ ,<sup>11,12</sup> the *EL2* center in GaAs,<sup>13-17</sup> the *A* center in Si,<sup>18,19</sup> and the *M* center in InP.<sup>20-23</sup> These defects are reversible and charge state dependent,<sup>19,21</sup> and the effects following the large lattice relaxation have been explained with the help of configuration coordinate diagrams. Unlike *DX* and *EL2* centers, the configurations of the *M* and *A* centers were directly observed by deep-level transient spectroscopy (DLTS).<sup>18-23</sup>

Recent studies<sup>24-26</sup> have shown new metastable defects in *n*-type GaAs, depending on the heat treatment and the biasing condition<sup>24,25</sup> or the growth condition.<sup>26</sup> These defects, not the *EL2* center, were directly observed by DLTS. Their configuration has been explained as the charge-state reaction kinetics which relies on the pairing of a native acceptor or defect complex and a shallow donor, similar to models<sup>20-23</sup> proposed for other metastable defects.

In this paper we report that a new deep level in *n*-type GaAs is generated during hydrogenation, giving rise to disappearance of native deep levels, and that the hydrogen atom plays an important role in the metastability of deep levels found in hydrogenated *n*-type GaAs. In addition, the metastable reaction kinetics and a possible identification of deep-level defects are discussed.

### II. EXPERIMENT

Si-doped GaAs bulk crystals with a carrier concentration of  $(1.2-1.6) \times 10^{17} \text{ cm}^{-3}$  were used as starting materials and were exposed to hydrogen plasma at 250 °C in a capacitively coupled rf plasma-enhanced chemical vapor deposition (PECVD) system. The rf frequency was 13.56 MHz. The hydrogen pressure during the discharge was 0.57 Torr with discharge power densities from 0.01 to 0.1 W/cm<sup>2</sup>. The unbiased anneal for the hydrogenated sample was done in a vacuum furnace from 220 °C to 300 °C and the biased anneal for the fabricated diode was carried out below 150 °C. Before hydrogenation, the samples were chemically etched ( $[\text{H}_2\text{SO}_4]:[\text{H}_2\text{O}_2]:[\text{H}_2\text{O}] = 6:1:1$ ) to a depth of 0.5  $\mu\text{m}$  from the surface in order to eliminate the effects of the mechanically damaged layers.

An Au Schottky diode with a diameter of 0.5 mm was prepared using a thermal evaporator. The activation energy, the capture cross section, and the concentration of each trap were obtained using a DLTS system controlled by a microcomputer.

### III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) represent the DLTS spectra of as-grown and hydrogenated samples, respectively, and Figs. 1(c)–1(e) correspond to a series of isothermal annealing times under a reverse bias 10 V. In the DLTS measurements, the rate window was fixed to  $46.21 \text{ s}^{-1}$  in all cases. In the as-grown samples (a) with  $N_d = 1.2 \times 10^{17} \text{ cm}^{-3}$ , two deep levels at 0.81 and 0.33 eV below the conduction band, denoted as the *EL2* and the *H4* traps, respectively, exist as typical native deep levels in the concentration range of  $10^{15} \text{ cm}^{-3}$ . As seen in Fig. 1(b), after hydrogenation, the concentrations of these two deep levels decrease rapidly and especially the *H4* ( $E_c - 0.33 \text{ eV}$ ) trap almost disappears. In this case, it was shown that two new deep levels are generated, which are denoted as the *H2* ( $E_c - 0.60 \text{ eV}$ ) and the *H3* ( $E_c - 0.42 \text{ eV}$ ) traps, respectively. Also, in another as-grown sample with the *EL2* trap and the *H3* trap (instead of the *H4* trap), the *H3* trap disappears during hydrogenation. More importantly, from the fact that the *H2* concentra-

tion is higher in the sample in which the *H4* concentration (or the *H3* concentration) is higher than that of the *EL2* trap, it is likely that these levels are strongly related to the metastable configuration. During  $150^\circ\text{C}$  isothermal annealing under 10-V bias, the concentration of the *H2*, the *H3*, and the *H4* traps varies greatly with the annealing time, while the *EL2* trap increases gradually with annealing. However, it is noted that the unbiased-annealed samples below  $150^\circ\text{C}$  did not show the concentration variations of these deep levels as well as shallow donor levels. The quoted activation energies for the *H2* and the *H4* traps are found to be slightly different from the *EN* traps in Ref. 10, which shows the broad DLTS signal with overlapped peaks.

The concentration variations of the *H2*, *H3*, and *H4* traps are plotted in Figs. 2 and 3 as a function of annealing time at  $100^\circ\text{C}$  and  $150^\circ\text{C}$ , respectively. Figure 2 shows the results of a series of isothermal anneal at  $100^\circ\text{C}$  under a 10-V reverse bias. The  $100^\circ\text{C}$  data represent the symmetrical behavior in their concentrations between the *H2* and the *H3* traps: The *H3* trap increases up to 50 min and then slowly decreases while the *H2* trap decreases and then increases. Also, the *H4* trap increases rapidly up to 50 min and decreases after 50 min. Then, the maximum concentration of the *H4* trap at 60 min reaches  $3.5 \times 10^{15} \text{ cm}^{-3}$ . The change of the *H2* concentration is approximately equal to that of the *H3* concentration. However, the changes of the *H2* and the *H3* concentrations are not associated with that of the *H4* concentration. By annealing at  $150^\circ\text{C}$  (Fig. 3), the *H4* and the *H3* traps that disappeared during hydrogen plasma exposure (HPE) are recovered for the first 10 min, but as the annealing time elapses up to 40 min, it is reduced to  $(5-10) \times 10^{14} \text{ cm}^{-3}$ . From 45-min annealing, however, it is recovered again and into the original quantity after 90 min. These data also confirm that the *H2* trap shows a perfect symmetrical behavior as compared with

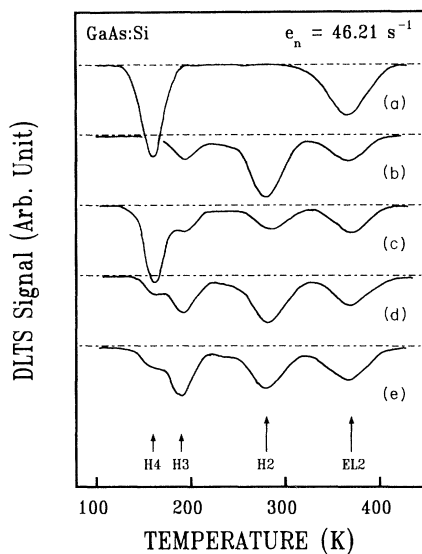


FIG. 1. DLTS spectra for the samples hydrogenated by hydrogen plasma exposure (HPE) at  $250^\circ\text{C}$  for 3 h. (a) is for the as-grown sample which shows only two native deep levels (the *EL2* and the *H4* traps) and (b) for the as-hydrogenated sample which shows a new level (the *H2* traps) with complete disappearance of the *H4* trap. The (c)–(e) figures are the results of a series of isothermal annealing at  $150^\circ\text{C}$  under a 10-V reverse bias. In the DLTS measurement, the rate window was fixed to  $46.21 \text{ s}^{-1}$  in all cases.

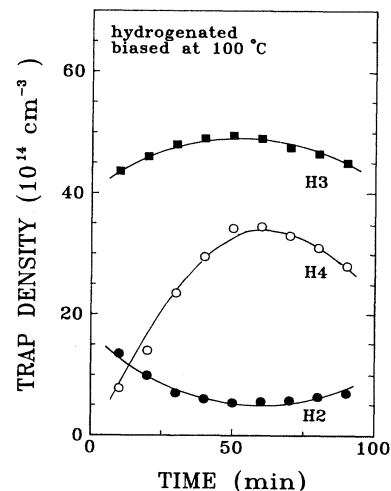


FIG. 2. Concentration of the *H2*, *H3*, and *H4* traps as a function of the annealing time at  $100^\circ\text{C}$ . This shows the result of a series of isothermal anneal at  $100^\circ\text{C}$  under a 10-V reverse bias.

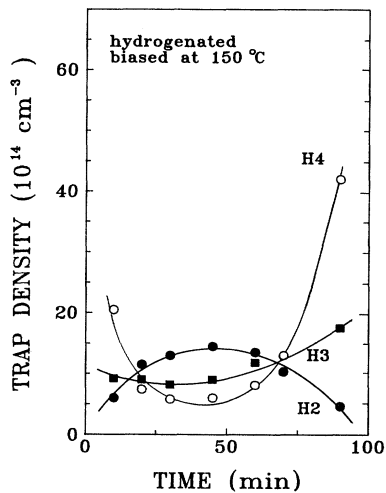


FIG. 3. Concentration variation of the deep-level defects with annealing time under 10-V bias. While the *EL2* and the shallow donors increase with the annealing time at 150°C, the *H3* and the *H2* traps show compensating behaviors in their concentrations and then recover near their original quantities.

that of the *H3* trap, i.e., the increase of the *H2* trap up to 40 min and the decrease after 45 min. The maximum concentration of the *H2* trap during the biased isothermal annealing reaches  $1.5 \times 10^{15} \text{ cm}^{-3}$  after 45 min when it appears as a major deep level. This result indicates that the *H2* trap generated during HPE is the hydrogen complex associated with the *H3* trap and that the hydrogen atom can be related to the presence of the metastable defect. Another deep level passivated during HPE, the *H4* trap, varies as the increase of the annealing time and has a generation frequency similar to the *H3* trap and shallow donor, but the *H4* trap does not reappear in the unbiased-annealed samples. Since the *H2* trap and the *H3* trap show a complete symmetrical behavior between their concentrations, we focus on these two traps.

To investigate more clearly the metastable behavior of the deep levels in the as-hydrogenated samples, the concentration profiles of the generated deep level (the *H3* and *EL2* traps were dominant) are drawn in Fig. 4 as a function of depth from the surface. The profiles were calculated from the DLTS measurements for chemically etched ( $[\text{H}_2\text{SO}_4]:[\text{H}_2\text{O}_2]:[\text{H}_2\text{O}]=6:1:1$ ) layers from the surface. The *H2* concentration, which is high in the donor-passivation region, within  $2.5 \mu\text{m}$  of the surface decreases with the increasing depth from the surface. However, the *H3* traps in the same region show opposite behavior in their concentration with the depth from the surface. The *H3* concentration increases into the original quantity above  $3 \mu\text{m}$  from the surface. In this case, the concentration of the passivated *EL2* trap recovers to  $1.5 \times 10^{15} \text{ cm}^{-3}$ . It is interesting to note that during the chemical etching for the *H3*-dominant and hydrogenated sample, we could not find the *H4* trap. This result indicates that the *H4* trap could be the defect regenerated by

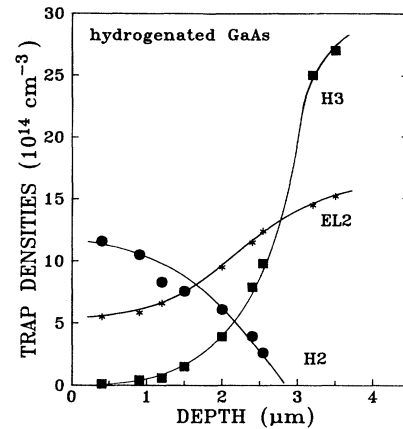


FIG. 4. Concentration profiles of the deep-level defects in as-hydrogenated Si-doped GaAs as a function of depth from the surface. The profiles were calculated from the DLTS measurement for chemically etched ( $[\text{H}_2\text{SO}_4]:[\text{H}_2\text{O}_2]:[\text{H}_2\text{O}]=6:1:1$ ) layers from the surface.

the applied electric field during the electric-field-enhanced anneal. From the results that the generated *H2* concentration is higher in samples with the high concentration of the *H3* trap, it is evident that the *H2* trap is associated with the *H3* trap and its metastable behavior is related to the atomic hydrogen. During hydrogenation, it is possible that a defect is generated by plasma-induced damage<sup>27</sup> during the bombardment as well as the exposure to atomic hydrogen. In order to elucidate whether these traps are defects induced from the possible plasma-induced damage, the effect of varying the discharge power density during hydrogenation was investigated. Figure 5 shows DLTS spectra for the sample hydrogenated with plasma power densities from 0.01 to 0.1  $\text{W}/\text{cm}^2$  at 280°C. The used sample was moderately doped ( $6 \times 10^{16} \text{ cm}^{-3}$ ) and the rf plasma frequency was 13.56 Mhz. The *H2* trap appears initially in the hydrogenated sample at a power density of 0.01  $\text{W}/\text{cm}^2$ . Its concentration increases up to 0.06  $\text{W}/\text{cm}^2$  power density and decreases gradually above 0.06  $\text{W}/\text{cm}^2$  power density, showing a maximum *H2* concentration of  $1.3 \times 10^{15} \text{ cm}^{-3}$  at 0.06  $\text{W}/\text{cm}^2$ . Considering the fact that the hydrogenation under a power density of 0.06  $\text{W}/\text{cm}^2$  gives the most effective passivation of Si donor in the Si-doped GaAs, it is thought that the *H2* generated during hydrogenation might be induced from hydrogen incorporation in Si-doped GaAs. Also, in Si-doped and undoped GaAs exposed by nitrogen and argon plasmas we could not observe the generated deep levels. Therefore, it is clear that the generated deep levels are due to hydrogen incorporation in Si-doped GaAs during hydrogenation. In fact, since the possibility of defects due to the plasma-induced damage cannot be excluded, further experimental studies should be done.

Assuming that the deep-level formation and annealing

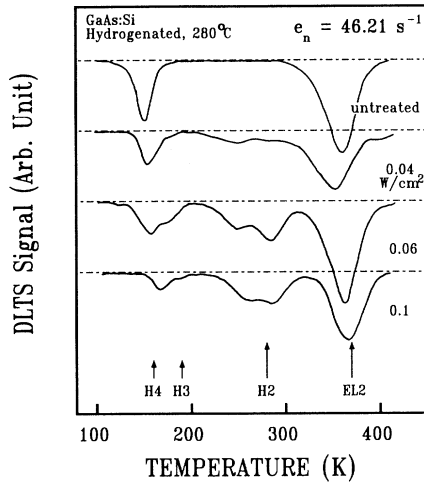


FIG. 5. DLTS spectra for the untreated sample and the hydrogenated samples exposed at 0.04, 0.06, and 0.1  $\text{W}/\text{cm}^2$ , respectively. The substrate temperature during hydrogenation was  $280^\circ\text{C}$ .

follow the first-order kinetics,

$$N(t, T)/N_0 = \exp(-\nu_a t) \quad (1)$$

and

$$N(t, T)/N_0 = 1 - \exp(-\nu_f t), \quad (2)$$

respectively, where

$$\nu = \nu_0 \exp(-E_a/kT). \quad (3)$$

Here  $t$  and  $T$  are the annealing time and temperature.  $N_0$  is the maximum deep-level concentration,  $E_a$  the binding energy, and  $\nu_f, \nu_a$  the deep-level formation and annealing frequencies, respectively. The formation frequencies  $\nu_f$  calculated from Eq. (2) are  $1.45 \times 10^{-4} \text{ s}^{-1}$  for the  $EL2$  trap,  $1.28 \times 10^{-4} \text{ s}^{-1}$  for the  $H3$  trap, and  $0.95 \times 10^{-4} \text{ s}^{-1}$  for the donor at  $150^\circ\text{C}$ . In these plots, it is confirmed that the  $H3$  and the  $H4$  traps are not newly generated deep levels, but native ones which appear dominantly after HPE because they are neutralized more slowly than other native deep levels during hydrogenation. The attempted frequencies  $\nu_f, \nu_a$  and the binding energy  $E_a$  of a deep level under the biased annealing condition show a large difference from those under unbiased conditions. This difference is due to the fact that the hydrogen atom can be electrostatically driven in GaAs by the electrical field. In order to investigate the electric-field-strength dependence of the hydrogen atom in the solid, the exact width of the space-charge region in the metal-semiconductor junction is to be known. This width increases generally by the increase of a reverse bias voltage  $V_r$ , and depends on the distribution of charged traps and impurities. Therefore, it is difficult to obtain exactly the required electric-field strength by only a reverse bias condition in the metal-semiconductor structure. For this reason, the experimental values such as the attempted

frequencies and binding energy can scatter according to the field strength. The first-order kinetics of the deep levels was considered for the samples isochronally annealed under the unbiased condition at temperatures in the range of  $220$ – $280^\circ\text{C}$ . Figure 6 represents the data corresponding to the  $H2$ , the  $H3$ , and the  $EL2$  traps. In the  $240^\circ\text{C}$  unbiased annealing condition, the calculated formation frequencies  $\nu_f$  are  $1.3 \times 10^{-2} \text{ s}^{-1}$  for the  $H4$  trap and  $2.4 \times 10^{-2} \text{ s}^{-1}$  for the  $EL2$  trap, and the annealing frequency  $\nu_a$  of the  $H2$  trap is  $1.3 \times 10^{-2} \text{ s}^{-1}$  at  $240^\circ\text{C}$ . Using Eqs. (1), (2), and (3), the activation energies responsible for the formation of the  $EL2$  and the  $H3$  traps and for the annealing of the  $H2$  trap can be extracted. The annealing energy of the  $H2$  trap is  $1.61 \pm 0.04 \text{ eV}$ ; the formation energies of the  $EL2$  and  $H3$  traps are  $1.88 \pm 0.05 \text{ eV}$  and  $1.59 \pm 0.05 \text{ eV}$ , respectively. Taking into account these results in which the annealing energy and the annealing frequency of the  $H2$  trap are consistent with the formation energy and frequency of the  $H3$  trap, it is confirmed that the metastable transformation between the  $H2$  and the  $H3$  traps can be thermally induced and controlled by an electric field, and that a hydrogen atom might be responsible for the complete metastable reaction kinetics of two configurations. The  $H2$  ( $E_c - 0.60 \text{ eV}$ ) and  $H3$  ( $E_c - 0.42 \text{ eV}$ ) traps are similar to metastable defects reported in metal-organic chemical vapor deposition (MOCVD) grown GaAs, in which defect configurations depend on the applied bias voltage during thermal annealing at  $400 \text{ K}$  (Refs. 24 and 25) and the  $[\text{As}]/[\text{Ga}]$  ratio during the growth.<sup>26</sup> The results of Buchwald *et al.*<sup>25</sup> and Tabata *et al.*<sup>26</sup> provided the estimate of formation and annealing energies associated with the metastable

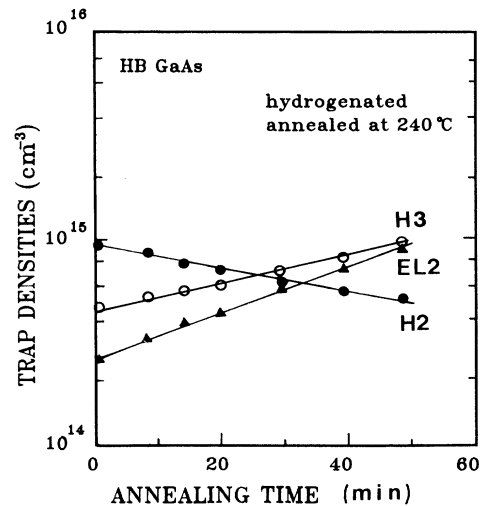


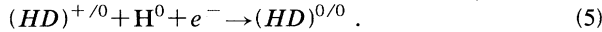
FIG. 6. Concentration variation of the deep levels with annealing time at  $240^\circ\text{C}$  under zero bias. The logarithmical changes of the  $H2$  and  $H3$  traps with the annealing time are different from those for the biased annealings. In these plots, the temperature-dependent annealing frequencies for the  $H2$  trap satisfy the relation  $\nu_a = 0.82 \times 10^{13} \exp[(-1.61 \pm 0.04 \text{ eV})/kT] \text{ s}^{-1}$ , and the activation energy of  $1.61 \pm 0.04 \text{ eV}$  is expected to be the energy required for the release of a hydrogen atom bound to a native defect.

configurations. The values  $1.70 \pm 0.08$  eV (Ref. 25) and  $1.8 \pm 0.2$  eV (Ref. 26) corresponding to the  $M$  trap (0.31 eV  $\rightarrow$  0.61 eV) and the  $E$  trap (0.33 eV  $\rightarrow$  0.56 eV) transformations, respectively, agree reasonably well with our experimental results. They<sup>26</sup> proposed that during the growth condition of MOCVD, isolated defects due to excess arsenic, such as arsenic interstitial  $As_i$  or arsenic antisite  $As_{Ga}$  can be candidates for the origin of the metastable defects in GaAs. Also, others<sup>25</sup> suggested a possible origin for a configurational transformation of deep levels in GaAs: Since the deep level has a large lattice relaxation and appears in the near surface region, it might be a stress-induced defect due to the nitride encapsulation or the nitride complex with a bulk impurity near the surface that might have diffused out during processing. The work of Jalil *et al.*<sup>27</sup> shows that the  $E_c - 0.55$  eV and the  $E_c - 0.41$  eV trap in the sample hydrogenated under a power density of  $0.1$  W/cm<sup>2</sup> are likely residual defects generated in the plasma-damaged region. Since the metastability of these traps was not reported, however, we cannot elucidate whether these traps are identical to the  $H2$  and the  $H3$  traps. Although the reported traps (the  $M$  traps,<sup>24,25</sup> the  $E$  traps,<sup>26</sup> and the work of Jalil *et al.*<sup>27</sup>) are similar to the  $H$  traps in respect of the formation and annealing energy and/or the activation energy, it is difficult to definitively elucidate whether the  $H$  traps are identical to those reported in Refs. 24–27.

In light of the above results, we propose a model which includes a multicharged defect  $(HD)^{+/+}$  and hydrogen atom  $H^0$  with the gain of electrons. The metastable configuration by a hydrogen atom,  $H^0$ , is expressed according to the reaction

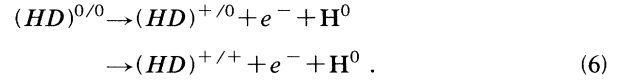


or as additional hydrogen atoms with electrons are supplied subsequently,



In Eqs. (4) and (5), assuming  $(HD)^{+/+}$  as the  $H3$  trap, and  $(HD)^{+/0}$  [or  $(HD)^{0/0}$ ] as the  $H2$  trap, it is suggested that the defect partially  $(HD)^{+/0}$  or fully  $(HD)^{0/0}$  passivated by a hydrogen atom may be the metastable defect for  $(HD)^{+/+}$  (the  $H3$  trap). In the  $H2$  configuration, it can be loosely bound to the nearest atoms because of a hydrogen atom, and then a hydrogen atom with electrons can be released easily by the electric field. Therefore, the

electrically driven or the thermally activated dissociations can also occur with the loss of electrons or hydrogen atom. This is to say the pair ( $H2$ ) is dissociated according to the relation



In addition, we propose a tentative atomic configuration of the  $H2$  and the  $H3$  traps. If the atomic configuration of the  $H3$  trap, as previously reported<sup>27–29</sup> by others (it will not be discussed here as it is beyond the scope of this paper) is  $(As_{Ga} - V_{As})^{+/+}$ , the identification of the  $H2$  trap can be considered as follows: During hydrogenation, the arsenic vacancy  $V_{As}$  or the arsenic antisite  $As_{Ga}$  of the above structure is passivated by a hydrogen atom, forming the partially passivated  $(As_{Ga} - V_{As})^{+/0}$  configuration. The partially or fully passivated  $(As_{Ga} - V_{As})$  configuration becomes a tentative candidate of the  $H2$  trap. However, for identification of the exact origins of these configurations, a more detailed theoretical study is required.

#### IV. SUMMARY

We have shown the role of the hydrogen atom on deep levels in GaAs. The deep level at  $E_c - 0.60$  eV is generated newly during hydrogenation and it is the metastable defect for the  $E_c - 0.42$  eV trap. The activation energies responsible for the formation and the anneal of these two level configurations are  $1.59 \pm 0.05$  eV and  $1.61 \pm 0.04$  eV, respectively. Also, it is believed that the 0.60-eV trap in Si-doped or undoped GaAs is a hydrogen complex partially or fully associated with the 0.42-eV trap. We conclude that the hydrogen atom plays an important role in the metastability of deep-level defects in GaAs and that the activation energy of  $1.61 \pm 0.04$  eV could be the energy required for the release of a hydrogen atom bound to arsenic vacancy  $V_{As}$ .

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