Charge-trapping model of metastability in doped hydrogenated amorphous silicon

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A new theory of metastability phenomena in doped hydrogenated amorphous silicon (*a*-Si:H) is described. It is proposed that an ensemble of bistable charge-trapping defects causes the metastability. It is demonstrated that in doped films there is a different dominant mechanism of metastable defect formation than in undoped films. Charge trapping is an essential component of defect formation in the doped films, and the new theoretical model uses this observation to unify the analyses of quenching, illumination, and depletion-bias annealing experiments. Bistable charge-trapping defects, such as the well-known *DX* center in $Ga_{1-x}Al_xAs$, have also been observed in many doped crystalline semiconductors. Specific defect reactions involving bond formation or bond switching to activate the dopant and explicitly including the observed charge trapping are proposed as the source of metastability in the doped *a*-Si:H. Some implications of this model for our understanding of light-induced effects in undoped *a*-Si:H are also discussed.

I. INTRODUCTION

The stability of hydrogenated amorphous silicon (*a*-Si:H) has been widely studied[1] for its scientific interest and because of the deletrious effects of illumination and carrier injection upon a variety of electronic devices, including photovoltaic cells and field-effect transistors. Most workers study light-induced effects in undoped or lightly doped *a*-Si:H, but several groups have examined light-induced effects in more heavily doped[2-6] films. Metastable effects due to the rapid quenching of doped films[7-9] and to the annealing under bias of doped layers in device structures[10-12] have also been observed.

In this paper it is shown that a bistable charge-trapping defect model accounts for the quenched-in, bias-annealed, and light-induced effects in doped *a*-Si:H and that the dominant defect reaction in phosphorus-doped *a*-Si:H is not observed when undoped *a*-Si:H is illuminated. I propose that the dominant defect reaction in the P-doped films involves hole trapping or electron emission that accompanies bond formation between a threefold-coordinated P and a neighboring atom or bond switching to the P. This activation of the P atom causes the Fermi level, E_F, to move toward the conduction-band mobility edge, E_c. The rehybridizations and atomic-position shifts present a significant barrier to reconfiguration in this bistable defect system. Specific proposals for the defect reaction in the P-doped films are presented and evaluated. Metastability in B-doped films and some aspects of light-induced degradation in undoped films are also discussed in light of the phenomenology of bistable charge-trapping defects.

Bistable charge-trapping defects have also been observed and studied in a variety of doped crystalline semiconductors. Examples include the iron-acceptor pair in Si:Al,[13] the *C* center (likely the boron-vacancy pair) in Si:B,[14] the interstitial-carbon–substitutional-carbon pair in both *n*-type and *p*-type Si,[15] the *DX* center in $Ga_{1-x}Al_xAs$,[16] the *M* center in *n*-type InP,[17] and the *M*Fe center in *n*-type InP:Fe.[18] Each of these defects is bistable because its stable configuration changes with the defect's charge state and a substantial barrier retards changes in configuration.

II. BACKGROUND: METASTABILITIES IN DOPED *a*-Si:H

Three different treatments of heavily doped *a*-Si:H have resulted in metastable increases in conductivity, Fermi-level position, and/or doping efficiency. The first treatment, rapid quenching of P-doped films, causes a metastable increase of conductivity that results from a motion of E_F toward E_c.[7-9] For the same reason, quenching increases the total charge swept out of the *n* layer of an *i*-*n*-*i* structure upon application of a voltage step.[11] The additional sweep-out charge lies between the initial and the quenched-in Fermi levels.[19]

The second treatment can be called depletion-bias annealing because it involves depleting the *n* layer of majority carriers at elevated temperature. For example, Street and Kakalios[11] annealed *i*-*n*-*i* structures at 130 °C under an applied bias that depleted the *n* layer of majority carriers. They cooled the sample under this depletion bias and found an increase in swept-out charge at room temperature comparable to that induced by quenching. These authors conclude that quenching and depletion-bias annealing of an *n*-doped layer are identical processes. There is a metastable increase of E_F after each treatment. Depletion-bias annealing has also caused metastable changes in *n*-type Schottky-barrier structures.[10,12] Lang *et al.*[10] annealed Schottky barriers in reverse bias at 200 °C, cooled them under reverse bias, removed the reverse bias, and measured capacitance-voltage curves at room temperature. The measurements revealed an increased density of band-tail electrons between $E_c - 0.5$ eV and E_F in those bulk regions that were depleted of electrons during the anneal. These authors conclude that depletion-bias annealing causes an increase in activated

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(fourfold coordinated) donor concentration that moves the Fermi level toward E_c and accounts for the increased density of band-tail electrons after the anneal.

The third treatment that causes metastable conductivity changes in heavily doped a-Si:H is prolonged illumination at elevated temperature.[2,20,21] Jang and Lee[20] report that the conductivity of P-doped dc-glow-discharge a-Si:H increased more than a factor of 2 after about 3 h exposure to 150 mW/cm^2 of (7000–9000)-Å light. It is reasonable to attribute this increase in conductivity to the movement of E_F toward E_c. Metastable light-induced increases in the conductivity of B-doped a-Si:H are also reported.[2,20,21] Jackson *et al.*[3] observed a photoinduced transparency (PT) in heavily-P-doped a-Si:H, which they attribute to electrons thermalizing into activated donors that have a lower optical cross section than that of band-tail states. The magnitude of this PT undergoes a metastable increase as the sample is illuminated. The authors attribute the metastable increase of the PT to an increased activated donor concentration caused by the illumination. In spite of a measured decrease in E_F that may be due to other defect reactions, Jackson *et al.*[22] also observed a light-induced increase of a hyperfine-split electron-spin-resonance (ESR) line, which they attribute to the occupied activated donor state. Since the decrease in E_F leads to a reduced occupation of the donor level, this result also indicates an increase in the concentration of ionized activated donors upon illumination.

If a single-defect-reaction mechanism underlies the similar metastable changes induced in P-doped films by quenching, depletion-bias annealing, and illumination, then the annealing kinetics for each of these changes should be the same. Table I exhibits the annealing activation energy E_a and the prefactor ν for metastability experiments reported in the literature. Although the metastable changes were produced by the three different treatments, the annealing data cluster around relaxation rates given by approximately $(10^8\ \mathrm{s}^{-1})\exp(-0.9\ \mathrm{eV}/kT)$, supporting the idea of a common mechanism.

One might suppose that the defect reaction driven by the quenching or depletion-bias annealing of doped films is the same as the defect reaction driven by illuminating undoped a-Si:H. Several observations suggest that this is not so. First, reversible quenched-in conductivity increases become increasingly difficult to observe as the P-doping level is decreased,[9] indicating that P may participate in the metastable changes. Second, heavily doped films have exhibited light-induced conductivity increases, while lightly doped films from the same system exhibited the usual conductivity decrease associated with undoped films.[2]

Third, the annealing activation energies and prefactors (see Table I) for metastable defects in P-doped films are smaller than the usual annealing energies for light-induced defects in undoped films. A more definitive distinction based on annealing kinetics can be made by considering the sign of the charge emitted during annealing of the defects. Using capacitance techniques, Crandall[12] has studied the change in net charge associated with creation of metastable defects in both undoped and P-doped a-Si:H. He finds that the dominant defect created during depletion-bias annealing of an n-type Schottky device emits an electron during formation and captures an electron upon annealing. This is expected because depletion-bias annealing removes electrons from the film to form the defect. The annealing activation energy is 0.87 eV and ν is $4\times10^7\ \mathrm{s}^{-1}$, typical of the values for P-doped a-Si:H shown in Table I. His capacitance studies of metastable defect formation is undoped a-Si:H reveal various defects that anneal with different activation energies. Upon annealing, some of the light-induced defects in undoped films capture electrons and some capture holes. Whenever an electron is captured during annealing of undoped a-Si:H, $1.4 < E_a < 1.8$ eV and $10^{15} < \nu < 10^{19}\ \mathrm{s}^{-1}$, much higher values than are observed in P-doped a-Si:H. Thus, Crandall finds an unmistakably higher barrier to any defect annealing accompanied by electron capture in undoped a-Si:H than to the metastable defect annealing in P-doped a-Si:H, which is always accompanied by electron capture. Based on these three

TABLE I. Activation energy and prefactor for annealing of metastable defects that raise the Fermi level of P-doped a-Si:H. For each reference, the defect-creation and -detection techniques are also shown. Branz *et al.* (Ref. 9) studied a-Si:H deposited by laser-induced chemical-vapor deposition; all others studied glow-discharge a-Si:H.

Reference	E_a (eV)	ν (s^{-1})	Creation	Detection
Ast and Brodsky[a]	0.87	1.9×10^8	quenching	conductivity
Lang *et al.*[b]	0.87	6.4×10^7	depletion bias	capacitance
Street *et al.*[c]	0.95	1×10^{10}	quenching	sweep-out
Crandall[d]	0.86	4×10^7	depletion bias	capacitance
Jang *et al.*[e]	0.9–1.0	10^9	illumination	conductivity
Branz *et al.*[f]	1.1	2×10^7	quenching	conductivity

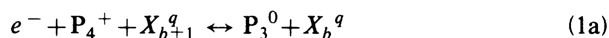
[a]Reference 7.
[b]Reference 10.
[c]Reference 23.
[d]Reference 12.
[e]Reference 2.
[f]Reference 9.

observations, I conclude that the dominant defect reaction in doped a-Si:H is different than any of the defect reactions that occur when undoped a-Si:H is illuminated.

A complicating factor is that the illumination of doped material can also result in formation of other metastable defects, most likely those that are formed by illumination of undoped material. Jang *et al.*[2] observe a conductivity increase upon illumination that anneals with E_a of 0.9–1.0 eV only in heavily-P-doped samples. It is this reaction that dominates most metastable effects in P-doped films and is the subject of this paper. However, their lightly doped samples exhibit a conductivity decrease that anneals with E_a of 1.3–1.5 eV, typical of undoped films. Upon illumination of heavily B-doped a-Si:H, Jang *et al.*[21] observed a rapid dark conductivity decrease followed by a slower increase. This suggests that illumination causes competing defect reactions in the B-doped film, with the slower one increasing the conductivity and dominating at long times. I focus in this paper on the reactions that *increase* the conductivity since these are also observed after quenching and depletion-bias annealing of both P- and B-doped a-Si:H.

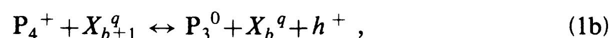
III. PROPOSED CHARGE-TRAPPING REACTION AND ITS IMPLICATIONS

To explain how these three distinct treatments of doped a-Si:H create the same defect, I propose that a pair of charge-trapping defect reactions take place at an ensemble of bistable defect sites. A bistable charge-trapping defect traps and stabilizes charges because each structural configuration of the defect has a different charge state and a substantial barrier retards changes in configuration.[24] To describe the defects and reactions proposed to explain the metastability in doped a-Si:H, I adopt the usual notation,[25] in which superscripts indicate the charge state of a defect, subscripts indicate its coordination number, and column-IV and -V atoms are denoted by T and P, respectively. The general form of the proposed bistable charge-trapping reaction is



$$e^- + P_4^+ + X_{b\pm1}^q \leftrightarrow P_3^0 + X_b^q \quad (1a)$$

and



$$P_4^+ + X_{b\pm1}^q \leftrightarrow P_3^0 + X_b^q + h^+ , \quad (1b)$$

where X could be a Si, H, or impurity atom, and q and b are integers. As reactions (1) go to the left, a hole is trapped or an electron emitted as X forms a bond to P (X_{b+1}) or loses a bond to P (X_{b-1}). Arrows to the right denote annealing and arrows to the left denote, for example, depletion-bias annealing with its accompanying increase in activated donor concentration. When reactions (1a) and (1b) proceed to the left, E_F increases. It is reasonable but not necessary[24] to assume that P is involved in the charge-trapping reaction and that the increase of E_F is associated with activation of P_3^0 to P_4^+. The assumption is supported by the PT and ESR measurements discussed earlier[3,22] and by the difficulty in observing these effects in undoped and lightly doped a-Si:H.[9] Reactions (1) are consistent with the capacitance experiments of Lang *et al.*[10] and Crandall,[12] which show that the defect becomes more positive when E_F increases.

Reactions (1) describe a bistable defect-complex system which is sketched in Fig. 1. The left-hand side is the upper state. When a bond forms between P and X, for example, the electronic states of both atoms rehybridize and there is a shift of atomic positions in the solid. P_3^0 is a purely p-bonded configuration with right angles between the bonds, but P_4^+ is sp^3-hybridized in a tetrahedral configuration. When the concentrations of each of the two configurations depart from equilibrium and an excess of P_4^+ is created, the shift of atomic positions presents a barrier of height, E_b, to reequilibration. Annealing of the metastable defects is activated with energy, E_b, as equilibrium is reattained.

Specific examples of X_b^q and the resulting reactions are discussed in Sec. IV, but I consider first the implications of reactions (1). The effect of quenching on a system with a defect population controlled by a bistable system was discussed at length in Ref. 9. When the temperature is above T^*, the fictive temperature for the defect reaction, the reaction is free to proceed and a temperature-dependent equilibrium concentration of the two defect configurations is reached. If the sample is then cooled below T^*, the equilibrium concentrations associated with T^* are frozen-in. Since T^* depends upon cooling rate[26] and is higher for rapid cooling, quenching freezes-in larger concentrations of the higher-energy defect configuration than does slow cooling. Therefore, annealing followed by quenching drives reactions (1) to the left and annealing followed by slow cooling drives them back to the right. This accounts for the observed conductivity changes.[7–9]

When a P-doped sample is held at a temperature above T^*, which depends on the annealing time,[26] the reactions proceed in both directions and equilibrium is maintained. Ordinarily, there is an equilibrium among reactions (1a), (1b), and electron-hole–pair creation and annihilation:

$$e^- + h^+ \leftrightarrow e^- h^+ . \quad (2)$$

Above T^*, both depletion-bias annealing and illumination are applied stresses which drive reactions (1) to the left. As described below, these are examples of

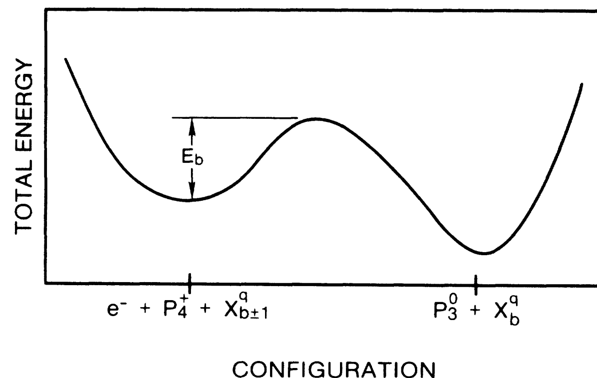


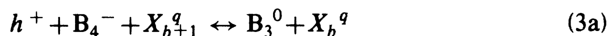
FIG. 1. Schematic diagram of the bistable system of defect configurations corresponding to reaction (1a). A barrier energy E_b retards equilibration of excess P_4^+.

LeChatelier's principle, which states that a system responds to applied stress to minimize that stress.

During depletion-bias annealing, electrons are drawn out of the material and reaction (1a) proceeds to the left in order to replenish the supply of electrons. The sample is cooled and the excess activated donors are frozen-in. This explains both the emission of electrons during depletion-bias annealing[12] and the increase of E_F found after cooling and removal of the bias.[10,11] From the thermodynamic viewpoint, the quasi-Fermi-level for electrons is lowered by the depletion bias, thereby reducing formation energies of positive defects and raising formation energies of negative defects.[27] The bias drives the equilibrium of reactions (1) to the left and cooling freezes-in excess concentrations of the more positive defect configuration. When the bias is then removed, E_F will have increased.

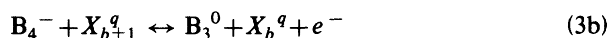
Upon illumination, nonequilibrium populations of free electrons and holes are created. Above T^*, illumination drives reaction (1b) to the left and reactions (1a) and (2) to the right, relative to the dark equilibrium. In n-type material, however, reaction (1b) dominates reaction (1a) for the reasons that follow. Under moderate illumination, we know that the fractional increase of the number of electrons in n-type material ($\Delta n / n$) is small because the photoconductivity of heavily doped a-Si:H is only slightly larger than the dark conductivity. The fractional increase of the number of holes ($\Delta p / p$) is much greater. Furthermore, in dark equilibrium, the high electron density and low hole density must drive reactions (1) to the right until suitable sites for the dopant deactivation reaction with P_4^+ and $X_{b\pm1}^q$ are nearly depleted. Adding more electrons, therefore, will not drive reaction (1a) much further to the right. However, the large increase of the number of holes during illumination drives reaction (1b) to the left and increases the active dopant concentration. This explains the increase in conductivity of P-doped films after illumination at elevated temperature.[2,20]

In a similar way, a pair of reactions of the form



$$h^+ + B_4^- + X_{b\pm1}^q \leftrightarrow B_3^0 + X_b^q \quad (3a)$$

and



$$B_4^- + X_{b\pm1}^q \leftrightarrow B_3^0 + X_b^q + e^- \quad (3b)$$

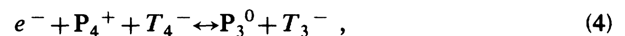
can account for the motion of E_F toward E_v observed when B-doped a-Si:H is quenched,[9,28] when i-p-i structures are depletion-bias-annealed[11,19] and when B-doped a-Si:H is illuminated at elevated temperature.[2,20,21] In this case, X is a Si, H, or impurity atom which forms a bond (X_{b+1}) or loses a bond (X_{b-1}) to B as an electron is captured or a hole is emitted.

Adler[29] and Crandall[12] have proposed that charge trapping into defect sites could also be responsible for the light-induced metastability observed in undoped a-Si:H and known as the Staebler-Wronski effect.[30] Crandall[12] used capacitance experiments to demonstrate that, when the Staebler-Wronski effect is produced by illumination or injection of carriers into undoped a-Si:H, electrons and holes are trapped and stabilized by defect reactions. As in reactions (1a) and (1b), each defect reaction causes the charge sign of the defect to change, but several factors complicate the analysis. First, Crandall[12] finds evidence of at least two different defects that undergo light-induced reactions. Second, in undoped material, illumination or double injection significantly enhance the populations of *both* electrons and holes because equilibrium concentrations of each are low. This drives each pair of reactions associated with a given defect in both directions and complicates the task of determining the dominant reaction in a given experiment. When a single sign of carrier is injected, it drives each pair of reactions in a unique direction, but under an applied voltage V only CV of charge can be single-injected into a device of capacitance C. Therefore, the stress applied to a given defect reaction in single injection is small and the resulting metastable change is also small.[12] If charge-trapping reactions cause the Staebler-Wronski effect in undoped a-Si:H, they would therefore be very difficult to study experimentally.

IV. SPECIFIC FORMS OF THE CHARGE-TRAPPING REACTIONS

I now speculate upon the specific forms of reactions (1) which dominate the metastable effects in P-doped films. During quenching, about 10^{17} cm^{-3} excess defects are frozen into the film.[9] This is a lower limit to the number of reaction sites for P_3^0 and X_b^q; there must be at least one such site for roughly every 10^5 Si atoms. Taking $[P]/[Si] = 10^{-2}$, and assuming that most P atoms are unactivated and that P_3^0 and X_b^q are randomly distributed, there must be at least one X_b^q per 10^3 atoms. The atom X could be an impurity species. However, lacking evidence of a correlation between the magnitude of these metastable effects and the concentrations of impurities other than P or B, I consider only Si defects and H for X_b^q. For brevity, I exhibit only the electron-emitting reaction that dominates the depletion-bias–annealing experiments.

Branz *et al.*[9] proposed the bond-formation reaction (X_{b+1})



$$e^- + P_4^+ + T_4^- \leftrightarrow P_3^0 + T_3^- , \quad (4)$$

without explicitly considering the charge trapping. The reaction is diagrammed schematically in Fig. 2. This re-

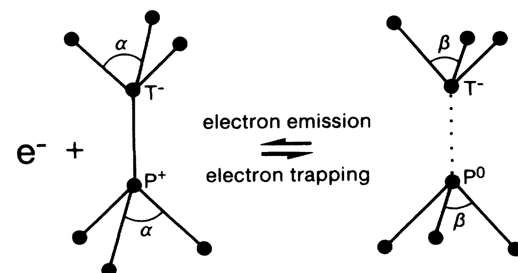
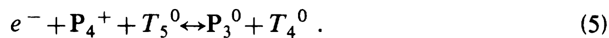


FIG. 2. Schematic diagram of reaction (4). Solid lines represent bonds. When the electron is emitted, a bond forms between the P and Si atoms. Angles α and β are approximately 110° and 90°, respectively.

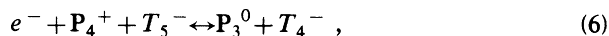
action involves only electron emission and bond formation between adjacent atoms and it promotes one electron to the conduction-band-tail state T_4^- and frees another, thereby raising E_F. After creation, the localized conduction-band-tail electron will migrate away from the P_4^+ and must make a random walk back before the defect can anneal by bond breaking. This could explain the low-annealing-rate prefactors observed for the metastable effect (Table I). Street *et al.*[31] find by a variety of techniques that the concentration of T_3^- in heavily-P-doped films is over 10^{18} cm^{-3}, so there should be enough sites at which T_3 can form a bond to P_3.

Two other possibilities involve formation of fivefold-coordinated Si, a defect suggested by Pantelides.[32] These reactions are diagrammed schematically in Fig. 3. Electron emission or hole trapping could induce bond formation between P_3^0 and T_4^0, according to



$$e^- + P_4^+ + T_5^0 \leftrightarrow P_3^0 + T_4^0 \ . \quad (5)$$

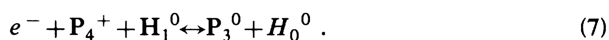
This reaction also changes E_F. There are obviously many possible sites for the bond formation, and the ease of migration of the fifth "floating" bond[32] is again consistent with the low-annealing-rate prefactors. However, the reaction requires that T_5^0 be stable in n-type a-Si:H, i.e., the T_5^- level lies above E_F. If T_5^0 is stable, then Pantelides's[32] assignment of the $g=2.0055$ ESR line to T_5^0 cannot be correct as the line disappears when doping raises E_F (Ref. 33) and the paramagnetic state is doubly occupied. Another possibility is



$$e^- + P_4^+ + T_5^- \leftrightarrow P_3^0 + T_4^- \ , \quad (6)$$

again involving bond formation between T_4^- and P_3^0. This reaction moves the Fermi level only if the T_5^- electronic states formed are at the Fermi level.

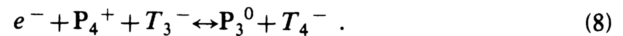
If X_b^q is a free hydrogen atom, H_0^0, the bond-breaking reaction would be



$$e^- + P_4^+ + H_1^0 \leftrightarrow P_3^0 + H_0^0 \ . \quad (7)$$

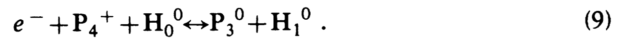
This defect reaction simply requires bond formation between an interstitial, unbonded H atom and a P_3^0 to increase E_F.

A bond-switching reaction (X_{b-1}) could be



$$e^- + P_4^+ + T_3^- \leftrightarrow P_3^0 + T_4^- \ . \quad (8)$$

This is diagrammed schematically in Fig. 4. Upon electron emission, a neighboring atom breaks its bond to the T_4^- atom with its localized conduction-band-tail electron and activates the P atom by bonding to it. A second bond-switching reaction could be



$$e^- + P_4^+ + H_0^0 \leftrightarrow P_3^0 + H_1^0 \ . \quad (9)$$

Reaction (8) appears similar to the reaction

$$P_4^+ + T_3^- \leftrightarrow P_3^0 + T_4^0 \quad (10)$$

proposed by Street[34] to govern the overall equilibrium between activated and unactivated P in doped a-Si:H. However, reaction (8) is a local reaction among nearby atoms, while Street *et al.*[35] envisage an overall equilibrium in the film mediated by hydrogen diffusion. Another difference is that as reaction (8) proceeds to the left, the emitted electron raises E_F, while reaction (10) raises E_F if and only if the T_3^- level created in the gap is at or above E_F.

With specific defect reactions in mind, the dependence of light-induced defect densities, ΔN, on doping and defect density can be examined. Skumanich *et al.*[4] used photothermal deflection spectroscopy (PDS) to find that ΔN is proportional to the gas-phase dopant concentration used in growing P-doped films. They find that the magnitude of ΔN is also proportional to total defect density in the doped films but independent of defect density in undoped films. The doping dependence of ΔN suggests that P is involved in the dominant defect reaction they are measuring. The dominant reaction they are measuring is likely the conductivity-increasing reaction discussed in this work. The number of sites undergoing each of the reactions (4)–(9) will increase with P concentration. As each reaction proceeds to the left, the density of PDS active gap states increases, and hence, ΔN is positive. The dependence of ΔN on the initial defect concentration in doped films is likely a consequence of the P-induced increase in defect states observed[31] in a-Si:H. In undoped a-Si:H, a different defect reaction that does not involve P dominates. Unlike the mechanism in doped films, the

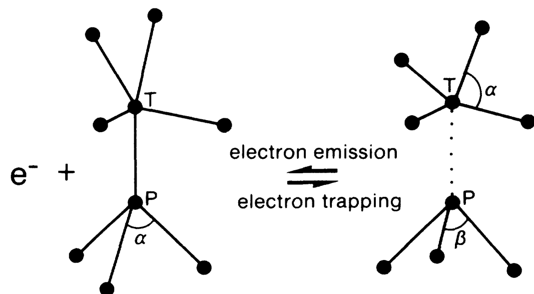


FIG. 3. Schematic diagram of reactions (5) or (6). Solid lines represent bonds. When the electron is emitted, a bond forms between the P and Si atoms. In reaction (5) the Si atom is neutral and in reaction (6) it is negatively charged. The angles α and β are approximately 110° and 90°, respectively.

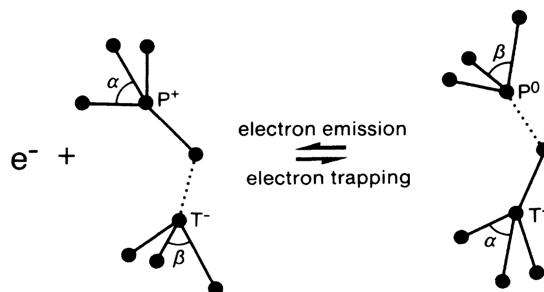


FIG. 4. Schematic diagram of reaction (8). Solid lines represent bonds. When the electron is emitted, the unlabeled atom switches its bond from Si to P. The angles α and β are approximately 110° and 90°, respectively.

number of reaction sites is evidently independent of initial defect concentration.

V. CONCLUSIONS

The detailed analysis of metastable changes in *a*-Si:H reveals that different mechanisms are dominant in doped and undoped films. In both P- and B-doped films, annealing under depletion bias or illumination results in movement of the Fermi level toward the nearest band-mobility edge, most likely due to an increase in active dopant concentration. This defect configuration can also be quenched into the doped films. Upon annealing P-doped films, a hole is emitted or an electron captured, but annealing energies and prefactors are far too small for identification with the hole-emitting annealing reactions observed in undoped *a*-Si:H after illumination.[12]

Hole trapping or electron emission are essential parts of the defect reaction observed when P-doped *a*-Si:H films are annealed under depletion bias or illumination and slow-cooled, or when they are quenched. The identity of the metastable changes induced by these three experiments was established and the common mechanism elucidated through a new theoretical model involving bistable charge-trapping defects. Specific defect reactions were discussed. These explicitly include the observed charge trapping and involve dopant activation by bond formation or bond switching.

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