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Amphoteric charge states and diffusion barriers of hydrogen in GaAs

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The states and microscopic diffusion mechanisms of hydrogen in crystalline GaAs are investigated by the *ab initio* molecular-dynamics method. The static equilibrium position for neutral hydrogen is found around the bond-centered (BC) site as in crystalline Si. As a negative U center, the hydrogen behaves as a stable donor around the bond-centered site in the p-type material, while it acts as a stable acceptor around the interstitial site surrounded by four nearest Ga host atoms in the n-type material. The hydrogen cation diffuses along the BC-C_{Ga}-BC-C_{As}-BC path in a high valence electron density region with a barrier of 0.46 eV in p-type materials. On the other hand, the hydrogen anion diffuses along the T_{Ga}-Hex-T_{As}-Hex-T_{Ga} path in a low electron density region with a barrier of 0.55 eV in the n-type material. The diffusion is more effective in p-type GaAs. Good agreement with experiments is found. Based on these results, the amphoteric behavior of hydrogen, its passivation effects of dopants, and its diffusion mechanisms can be understood in both p-type and n-type GaAs. [S0163-1829(99)02107-4]

Hydrogen has long been recognized as an all-purpose passivant for defects. The passivation of shallow acceptors and donors by hydrogen was first discovered in Si.[1] The incorporation of hydrogen in doped Si will deactivate the dopants and thus degrade the doped layers. It has also been indicated that the passivation is observed as a result of formation of neutral hydrogen-dopant complexes in Si.[1–3] The properties and effects of hydrogen in GaAs are similar to those of hydrogen in Si, namely, the passivation of dopants in doped GaAs. The passivations of both n-type and p-type GaAs were experimentally reported.[1,4–8] All of the donors Si, Ge, Sn, S, Se, and Te and the acceptors Be, Mg, Cd, Zn, and C are passivated by hydrogen.[9] The passivation depth is inversely dependent on the dopant concentrations. The thermal dissociation energy of the dopant-H complexes was obtained for several donors: $\mathrm{Si_{Ga}}$ (1.20 eV), $\mathrm{Sn_{Ga}}$ (1.20 eV), $\mathrm{Se_{As}}$ (1.52 eV); and for several acceptors: $\mathrm{Zn_{Ga}}$ (1.3 eV), $\mathrm{Si_{As}}$ (1.45 eV), C_{As} (1.35 eV), $\mathrm{Be_{Ga}}$ (1.15 eV), and $\mathrm{Cd_{Ga}}$ (1.35 eV),[10] where a symbol A_B indicates a substitution of B by A. The donor Si has been the most noticeable passivation impurity in GaAs.[8,11,12] The vibrational absorption of the $\mathrm{Si_{Ga}}$-H complex had been found and analyzed using Fourier transform infrared (FTIR) spectroscopy at the temperature between 5.5 and 300 K.[13] Among the acceptors, Be has a lower dissociation energy than Si. The low dissociation energies suggest that the passivation of acceptors and donors by hydrogen in GaAs are equally important. Technologically, hydrogen passivation has been employed in device fabrications, such as hydrogenated FET (HFET), semiconductor lasers, and optical waveguides, etc.

In the growth of GaAs layers, hydrogen plays a more decisive role than it does in Si. The hydrogen is invariably existent within the source chemicals, e.g., $\mathrm{AsH_3}$, or TBA's (tertiarybutylarsine) in which the bonded hydrogen can essentially suppress the incorporation of carbon acceptor in the GaAs layers.[14] In vapor phase epitaxy, $\mathrm{H_2}$ is usually used as the carrier gas for metalorganics, e.g., $\mathrm{Ga(CH_3)_3}$, to enhance the growth rate. Because of the technological importance of GaAs layers in high-speed microelectronic and optoelectronic devices, considerable attention has been paid to the role of hydrogen in crystalline GaAs.

In this paper, we present theoretical studies on the behavior of hydrogen as it diffuses through a GaAs crystal. From the fundamental aspect, it reveals the interaction between hydrogen and the prototypical III-V compound semiconductor. By changing the position of the Fermi level in the band gap, we can determine the stable sites and charged states for an isolated hydrogen. The results give us information about the effects of interaction between hydrogen and the dopants while they are well separated. Furthermore, the diffusion paths of hydrogen can be examined by our simulated diffusion along a specific trajectory.

The states of hydrogen in crystalline GaAs have been theoretically investigated earlier. Maric *et al.* indicated that the hydrogen prefers to occupy the position near the bond-centered site between a Ga and As atom with the Ga-As bond relaxing by 34.5% in the cluster $\mathrm{HGa_4As_4H_{18}}$.[15] Briddon and Jones noted that the hydrogen finds itself stable at the bond-centered site with a Ga atom substituted by an acceptor, while at the donor antibonding site a hydrogen-donor pair is formed in the cluster $\mathrm{Ga_{16}As_{26}H_{50}}$.[16] The similar configurations with hydrogen-donor or hydrogen-acceptor pairs were also found in the calculations on a periodic supercell with 16 and 32 host atoms of GaAs by Pavesi and Giannozzi.[17] The configuration is different from hydrogen in n-type crystalline Si with donors, where hydrogen is at the Si antibonding site. Pavesi and co-workers had also reported the amphoteric behavior of the hydrogen, based on the same supercells of 16 and 32 host atoms.[18,19] We have noted that the differences among the total energies corresponding to various hydrogen positions are of the order of tens of eV, ranging between 0.01 and 0.29 eV in the results of Pavesi and co-workers.[18,19] The small size of supercell may cause significant interactions between H atoms in neighboring supercells. It resulted in dispersion of the hydrogen level and placed deviations in energy calculations besides the well-known underestimation of the band gap by the local-density approximation for the electronic exchange-correlation functional. Furthermore, the relaxation of the host GaAs atoms around hydrogen may also be spatially extended in a larger supercell. The energy between different sites can have variations of 0.4 eV when a 32-atom cell is replaced by a 16-atom cell.[18,19] A relatively large supercell is needed to reduce the uncertainty.

The present calculations are performed using the Car-Parrinello (CP) molecular-dynamics methodology.[20] The valence electrons are described by density-functional theory in the local-density approximation (LDA). The norm-conserving electron-ion pseudopotentials[21] are used and modified to avoid a Ga ''ghost'' state.[22] For Ga and As atoms, the potentials are tested to successfully reproduce bulk structural properties.[23] The electronic wave functions are expanded on a plane-wave basis with a kinetic energy cutoff of 14 Ry (~9000 plane waves). The k points used in the first Brillouin zone (BZ) to represent the charge density are those that fold into the Γ point of the simple cubic supercell of 64 host atoms. It has been demonstrated that the Γ-point sampling in the BZ at such supercell size can effectively reproduce the static and dynamic properties of semiconductors.[24] Besides, in defect energy calculations, the Γ-point sampling is also successfully used in references 23, 25, and 26. Finally, no symmetry restrictions are imposed on the system during the calculations.

For atomic relaxation when any impurity or defect is introduced in the supercell, we use a more efficient scheme to obtain a stable structure. The scheme is still in the spirit of the CP method, where the electronic variables obey Newtonian equations to closely follow the ionic motions. While atoms are undergoing the normal molecular-dynamics evolution according to the Hellmann-Feynmann forces on them, a frictionlike force is introduced to slow down the atom moving upward on the Born-Oppenheimer surface. More specifically, the velocity of each atom is decomposed according to the direction of force acting on it. The component perpendicular to the force is eliminated. The parallel component is checked then. If it has the same sign as the force, it stays. Otherwise, the component is reduced by a factor of 3. In our experiences, this procedure is much faster in atomic relaxation than a simple steepest-descent or conjugate-gradient technique. Using this technique, we have obtained thorough relaxation for the system.

In our investigation on the hydrogen diffusion barriers, we adopt the so-called ''adiabatic trajectory'' simulation.[23] Along a specific diffusion path, the diffusing species, which is the hydrogen anion or cation, moves with a constant and small speed. The remaining atoms in the system relax simultaneously in the same scheme as described in the previous paragraph. The excess energy introduced by the constant speed motion of the diffusing atom is removed in the relaxation. The energy surface along the diffusion path is then probed.

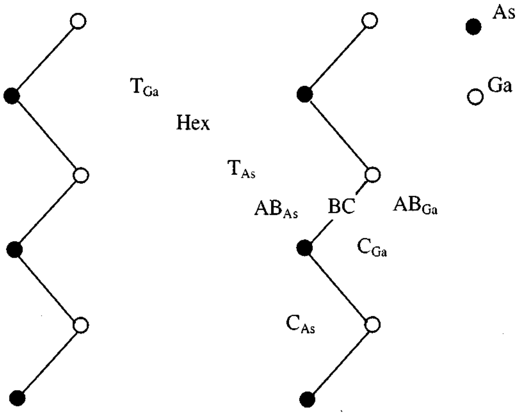

FIG. 1. Schematic illustration of the (110) plane through the atoms in GaAs crystal. The high-symmetry interstitial sites for H atom are denoted as follows. BC is the bond center, T the tetrahedral site (the subscripts indicate the first-nearest-neighbor ion), AB the antibonding site, C the site defined as the center of the rhombus formed between three adjacent ions and the nearest T sites, and Hex the hexagonal site. The M site is not on the (110) plane but lies in the middle of the axis connecting a BC with the nearest Hex site.

The stable sites and charged states for atomic hydrogen interstitials are determined by examining the total crystal energy at several symmetric positions in GaAs (see Fig. 1). They include a tetrahedral site surrounded by four nearest Ga hosts (T_{Ga}), a tetrahedral site surrounded by four As nearest hosts (T_{As}), an antibonding site nearest to a Ga host (AB_{Ga}), an antibonding site nearest to an As host (AB_{As}), a center site of the rhombus formed between three nearest Ga atoms and the T_{As} site (C_{Ga}), a center site of the rhombus formed between three nearest As atoms and the T_{Ga} site (C_{As}), bond-centered site (BC), hexagonal site (Hex), and M site. The M site is not on (110) plane. It is located midway on the axis connecting the BC with the nearest Hex. The calculated energies for various configurations of neutral hydrogen interstitials are listed in Table I, among which the energy of BC configuration is taken as zero. Our results indicate that the BC configuration has the lowest crystal energy as in Si. The stability of the neutral configurations goes in the order as follows: BC, C_{Ga}, AB_{As}, T_{Ga}, M, C_{As}, AB_{Ga}, T_{As}, and Hex. The energy difference between C_{Ga} and BC is 0.14 eV, while the energies of C_{Ga} and AB_{As} are degenerate. Hence, a neutral hydrogen atom in GaAs finds the stable position at the BC site, instead of the antibonding site of AB_{As} as pre-

TABLE I. Total energy differences for neutral H in GaAs with full relaxations. The zero of the energy is the global minimum for the relaxed crystal. The different columns correspond to the results for H in the different high-symmetry interstitial sites. BC is the bond center, T the tetrahedral site (the subscripts indicate the first-nearest-neighbor ion), AB the antibonding site, C the sites defined as the center of the rhombus formed between three adjacent ions and the nearest T sites, Hex the hexagonal site, and M the site in the middle of the axis connecting the BC with the nearest Hex site. The last row cites the results from the calculations of Pavesi, Giannozzi, and Reinhart (Ref. 18).

Site	BC	C_{As}	C_{Ga}	AB_{As}	AB_{Ga}	T_{As}	T_{Ga}	M	Hex
Energy (eV)	0.0	0.37	0.14	0.14	0.07	0.47	0.18	0.34	0.43
Ref. 18	0.19	0.21	0.11	0.0	0.23	0.22	0.29	0.11	0.25

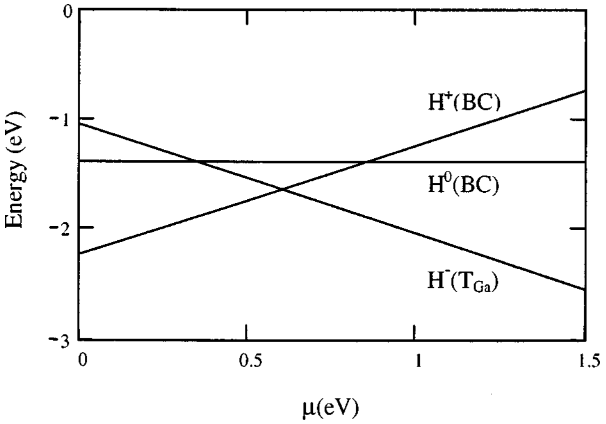

FIG. 2. Energies of various charge states of the interstitial hydrogen at their corresponding stable sites (in parentheses), relative to the neutral hydrogen in vacuum, as a function of the Fermi-level position μ in the gap.

viously predicted by Pavesi and co-workers.[18,19] Our results benefit by a larger defect spacing due to periodic boundary conditions and more thorough relaxation for atoms. The binding energy of a neutral hydrogen atom in the equilibrium BC site, which is obtained by comparison of the energies of the interacting system with those for the isolated H_2 and pure GaAs, turns out to be 1.39 eV (cf. 1.27 eV by Pavesi and co-workers).

The formation energy of a hydrogen interstitial is influenced by its charged state, which in turn depends on the Fermi-level position μ in the band gap. If the interaction between hydrogen and dopant can be ignored, the formation energy of a hydrogen interstitial in the charge state N is calculated as $E(N)-\mu N$, where N is the net electrons and $E(N)$ is the energy referred to the top of the valence band.[27] The stable charged state of a hydrogen interstitial could then be calculated at any Fermi level position μ. As the Fermi level changes in the band gap, the hydrogen finds for itself a stable charge state. In the region of high electronic charge density, the hydrogen interstitials tend to behave like donors. On the other hand, in the region of low electronic charge density, the hydrogen interstitials tend to behave like acceptors.

In the *p*-type materials, the hydrogen interstitial finds itself most stable in a positively charged state at the BC site. The compensation by a free hole exothermically occurs between a H^+ and an acceptor. In *n*-type materials, the hydrogen interstitial finds itself most stable in a negatively charged state at the T_{Ga} site. The compensation by a free electron exothermically occurs between a H^- and a donor. These results are in agreement with the hydrogen passivation effects, in which a passivated dopant is bonded to the charged hydrogen to form neutral dopant-hydrogen complex and annihilate a free carrier.

TABLE II. The relative energies among the various sites in high valence charge density region for H^+ in *p*-type GaAs. The zero of the energy is set at the global minimum.

Site	BC	C_{As}	C_{Ga}	AB_{AS}	M	T_{Ga}
Energy (eV)	0.0	0.35	0.44	0.62	0.870	1.23

TABLE III. The relative energies at several sites in low charge density region for H^{-1} in *n*-type GaAs. The zero of the energy is set at the global minimum. For comparison, the value at BC is listed as well.

Site	T_{Ga}	T_{As}	Hex	BC
Energy (eV)	0.0	0.29	0.55	0.89

Our results of calculations give us the information that single hydrogen interstitial in GaAs is a negative U center. The zero energy of the hydrogen interstitial is set at the neutral hydrogen in vacuum. As shown in Fig. 2, when the Fermi level moves from the top of valence band to edge of the conduction band, the stable hydrogen interstitial changes from H^+ to H^- state. The neutral hydrogen has never been a stable species. The similar conclusion has also been obtained for hydrogen as an impurity in crystalline silicon.[28] It should be noticed that associated with the uncertainty in LDA energy levels there is a possibility of the existence of H^0 near the midgap. The relative energy difference among the various sites in high valence charge density region for H^+ in *p*-type GaAs and in the low charge density region for H^- in *n*-type GaAs are shown in Tables II and III, respectively.

Voids surrounded by stacking faults, microtwins, and dislocations have been observed near the projected range in a high dose hydrogen-implanted GaAs sample using high-resolution transmission electron microscope at 550–600 °C by Sadana *et al.*[29] The phenomenological explanation rests on the forming of H_2 molecular bubbles in the GaAs. The hydrogen first precipitates until the H_2 molecules are nucleated. As the temperature is raised, they grow rapidly and burst to create and enlarge the surrounded defects. We have also carried out the calculations on the formation energy of a H_2 molecule. The H_2 molecule is the most favorable structure among hydrogen clusters H_n. Our calculations point out that the T_{Ga} site is the most stable position for H_2 (see Table IV). The energy for the molecule orientation along either the [100] or the [111] directions is almost degenerate. The calculated bond distance in the [100] and [111] direction is, respectively, 0.829 and 0.823 Å, which are slightly larger than that of an isolated hydrogen molecule in our calculation (with a value of 0.792 Å, to be compared with the experimental value of 0.75 Å). The energy of H_2 in the T_{Ga} site is higher than an isolated H_2 by 0.66 eV, but lower than that of a pair of two far separate neutral hydrogen atoms by 5.53 eV (cf. 0.5 and 5.6 eV, respectively, by Pavesi and co-workers). The energy of the H_2 structure is also lower than any other H_2^* complexes.[30] It implies that the H_2 molecules are nucleated through the hydrogen precipitating in GaAs.

Hydrogen has been known to be diffusive in bulk crystalline GaAs. Several experiments have described the hydrogen

TABLE IV. Relative energies (in eV) of H_2 in the interstitial sites of GaAs in both [100] and [111] orientation.

Site	T_{Ga}	T_{As}	BC
[100]	0.0	0.19	1.03
[111]	0.02	0.09	2.13

diffusion to be dependent on temperature, hydrogen concentration, and dopant concentration. The investigation of the diffusion profiles of hydrogen and dopant reveals that hydrogen diffuses mostly in atomic form at high temperature.[31] Molecular hydrogen is less mobile. The diffusing species could be neutral hydrogen, proton or H^-, depending on the doping in GaAs. Experimentally, diffusion is always greater in *p*-type GaAs for similar doping levels.[32] Therefore, the diffusion mechanism could vary with the Fermi level determined by doping.

We use the adiabatic trajectory scheme described previously to determine the migration barrier. We consider *p*- and *n*-type GaAs separately. As discussed earlier, in *p*-type GaAs where the Fermi level is close to the top of the valence band, H^+ is the most stable state. The global minimum for H^+ is at BC site. From one BC to a nearest-neighboring BC, we assign and maintain a small constant speed to a H atom in the direction connecting the two BC's. During the adiabatic trajectory simulation, the H atom is allowed to relax in the plane normal to the direction. It leads us to the saddle point of the energy surface when reaching the peak of the trajectory. The saddle point is found to be either C_{Ga} or C_{As}, depending whether the center atom between the two BC's is As or Ga. The energy barrier is 0.46 eV at C_{Ga}, and 0.36 eV at C_{As} [see Figs. 3(a) and 3(b)]. The lowest energy diffusion path between BC sites is hence in the high valence charge density region, i.e., BC-C_{Ga}-BC-C_{As}-BC, etc., with an activation energy of 0.46 eV. The path is similar to the one suggested for H^+ in *p*-type Si.[2,3,28,29] The barrier from BC to the low charge density sites is much higher. As can be seen in Table II, the energy for H^+ at T_{As} is 1.32 eV higher than that at BC. Therefore, H^+ is unlikely to migrate into the low valence charge density channel. Experimentally, an activation energy of 0.37 eV was found for jumping of H from a BC to another about a Be acceptor in *p*-type GaAs.[33] Our result is in good agreement with this measurement. But it needs to be noted that in our calculations H and the dopants are well separated. It should cost a higher energy when diffusing away from the acceptor in the case of the experiment. Indeed, the activation energy was estimated as 0.58 eV in a later experiment on deuterium diffusion profiles in zinc-doped GaAs.[34]

On the other hand, in *n*-type GaAs where the Fermi level is close to the bottom of the conduction band, the global minimum for H^- is at T_{Ga} site. The energy for H^- at T_{As} is only slightly higher than that at T_{Ga}. The energy barrier between these two sites occurs at the Hex site, as shown in Fig. 3(c). The lowest diffusion path for H^- is hence in the low valence charge density region, as described by a T_{Ga}-Hex-T_{As}-Hex-T_{Ga} path with an energy barrier of 0.55 eV from T_{Ga} to Hex as shown in Fig. 3(c). Since the energy cost for H^- from T_{Ga} to the BC site is higher (0.89 eV), H^- is unlikely to migrate into the high valence charge density region. By comparison with experiments, earlier experiments on the diffusion of H in *n*-GaAs got a spread of activation energies, from 0.62 (Ref. 35) to 2.2 eV.[36] A recent experiment on the diffusion of negatively charged muonium, which behaves like a light isotope of H, in *n*-GaAs, obtained an activation energy of 0.73 eV.[37] Since the energy surfaces we obtained are at the zero temperature, the mass of H does not affect the barriers.

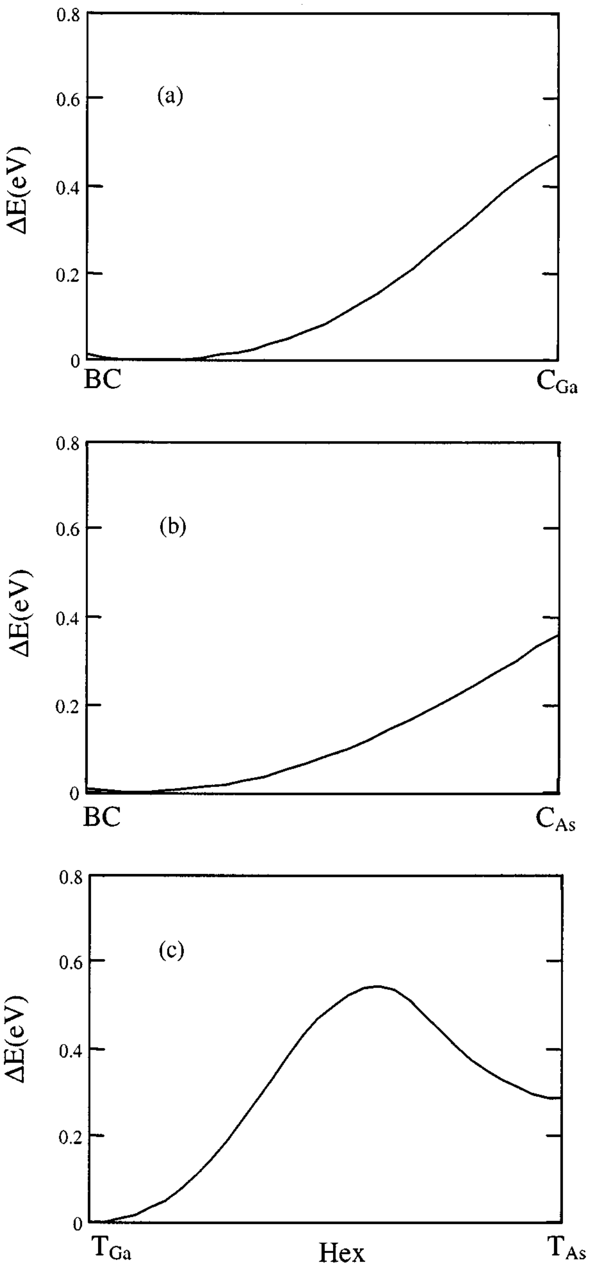

FIG. 3. Total-energy changes for the motion of H^+ along BC-C_{Ga} trajectory (a), H^+ along BC-C_{As} trajectory, and H^- along T_{Ga}-Hex-T_{As} trajectory (c). The same notations as in Fig. 1 have been used. In (a) and (b), the other half of the trajectories is not shown. It can be obtained by symmetry. The energy barrier for each trajectory is the maximum of the curve.

In summary, we have presented a microscopic description of hydrogen behavior when it diffuses through the crystalline GaAs by means of *ab initio* molecular-dynamics simulation. Our calculations suggest that hydrogen behaves as a donor in *p*-type GaAs or an acceptor in *n*-type GaAs. The hydrogen prefers diffusing through GaAs in atomic form. In *p*-type GaAs, the atom loses an electron to Fermi level and has its global minimum at the bond center. H^+ diffuses in the high valence charge density region. While in *n*-type GaAs, hydrogen atom acquires an electron from Fermi level and is most stable in the tetrahedral interstitial site surrounded by nearest Ga atoms. H^- finds the diffusion path in the low charge density region. The activation energies obtained in both cases are in good agreement with experiments. It is also consistent with the fact that atomic hydrogen is more diffuse in *p*-type than in *n*-type GaAs.

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