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Assessing the effect of hydrogen on the electronic properties of 4H-SiC

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As a common impurity in 4H silicon carbide (4H-SiC), hydrogen (H) may play a role in tuning the electronic properties of 4H-SiC. In this work, we systemically explore the effect of H on the electronic properties of both n-type and p-type 4H-SiC. The passivation of H on intrinsic defects such as carbon vacancies (V_C) and silicon vacancies (V_{Si}) in 4H-SiC is also evaluated. We find that interstitial H at the bonding center of the Si–C bond (H_i^{bc}) and interstitial H at the tetrahedral center of Si (H_i^{Si-te}) dominate the defect configurations of H in p-type and n-type 4H-SiC, respectively. In n-type 4H-SiC, the compensation of H_i^{Si-te} is found to pin the Fermi energy and hinder the increase of the electron concentration for highly N-doped 4H-SiC. The compensation of H_i^{bc} is negligible compared to that of V_C on the p-type doping of Al-doped 4H-SiC. We further examine whether H can passivate V_C and improve the carrier lifetime in 4H-SiC. It turns out that nonequilibrium passivation of V_C by H is effective to eliminate the defect states of V_C, which enhances the carrier lifetime of moderately doped 4H-SiC. Regarding the quantum-qubit applications of 4H-SiC, we find that H can readily passivate V_{Si} during the creation of V_{Si} centers. Thermal annealing is needed to decompose the resulting V_{Si}–nH (n = 1–4) complexes and promote the uniformity of the photoluminescence of V_{Si} arrays in 4H-SiC. The current work may inspire the impurity engineering of H in 4H-SiC.

Keywords: 4H-silicon carbide, hydrogen, electronic properties, passivation

PACS: 61.82.Fk, 61.72.–y, 71.15.Mb **DOI:** 10.1088/1674-1056/ac4234

1. Introduction

As a leading third-generation semiconductor, silicon carbide (SiC) is leaping in an explosive development to meet the increasing demand of electrical vehicles, 5G communications and renewable-energy systems. There exist over 250 polymorphs for SiC, among which 4H-SiC has attracted great attention owning to its wide bandgap, high carrier mobility, high thermal conductivity, and high stability.[1–3] 4H-SiC ingots are usually grown by the physical vapor transport (PVT) method, during which hydrogen gas (H_2) is used to optimize the surface of the seed crystal and tune the growth condition of 4H-SiC.[4–7] 4H-SiC epilayers are often homoepitaxially grown by chemical vapor deposition (CVD), during which H_2 is widely used as the carrier gas.[8–11] It should be also noted that H impurities are frequently incorporated during the processing of 4H-SiC-based devices.[12–15] Therefore, understanding the effect of H on the electronic properties of 4H-SiC is critical to the development of 4H-SiC technologies.

Considerable efforts have already been devoted to investigate H in 4H-SiC.[16–30] For n-type 4H-SiC, it was found that H–ion implantation, H_2-plasma treatment, and high-temperature H_2 annealing could reduce the concentration of electrons and increase the resistivity of 4H-SiC.[20–22] The reduction of the electron concentration was controversially attributed to the formation of N_C–H complexes, as well as the creation of electron traps such as H interstitials and intrinsic defects.[20] The hole concentration of p-type 4H-SiC also decreased after high-temperature H_2 annealing,[22–25] as a result of H_2 annealing-induced decomposition of Al_{Si}–H complexes and the creation of positively charge H,[22–24] which differed from the behavior of H in n-type 4H-SiC. H_2-plasma treatment was found to reduce the photoluminescence of both Al-related defects and N–Al complexes.[25] But the underlying mechanism remains ambiguous. Moreover, intrinsic carbon vacancies (V_C) were found to seriously reduce carrier lifetime in 4H-SiC.[26–29] It was experimentally found that H_2-atomsphere annealing was actually not capable of passivating V_C in 4H-SiC.[32,33] However, it was theoretically proposed that H might passivate V_C.[34–36] It is apparent that a clear picture on H in 4H-SiC remains elusive.

In this work, we systemically explore the effect of H on the electronic properties of both n-type and p-type 4H-SiC. The passivation of H on intrinsic defects such as V_C and silicon vacancies (V_{Si}) in 4H-SiC is also evaluated. We find that

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interstitial H at the bonding center of the Si–C bond (H_i^{bc}) and interstitial H at the tetrahedral center of Si (H_i^{Si-te}) dominate the defect configurations of H in p-type and n-type 4H-SiC, respectively. For n-type 4H-SiC, the compensation of H_i^{Si-te} is found to pin the Fermi energy and hinder the increase of electron concentration in highly N-doped 4H-SiC. Reducing the concentration of unintentional incorporated H is highly desired to improve the n-type doping and reduce the resistivity of n-type 4H-SiC. In p-type 4H-SiC doped by Al, we find that the compensation of H_i^{bc} on the p-type doping of 4H-SiC is negligible compared to that of V_C. Nonequilibrium H-incorporation approaches may passivate Al_{Si} by H, and degrades the performance of p-type 4H-SiC. The degradation can be easily eliminated by thermal annealing, because of the low binding energy of Al_{Si}–H in p-type 4H-SiC. We have further examined whether H can passivate V_C and improve the carrier lifetime in 4H-SiC. It turns out that nonequilibrium passivation of V_C by H is effective to eliminate the defect states of V_C, which enhances the carrier lifetime of moderately doped 4H-SiC. Regarding the quantum-qubit applications of 4H-SiC, we find that H can readily passivate V_{Si} during the creation of V_{Si} centers. Thermal annealing is needed to decompose the resulting V_{Si}–nH (n = 1–4) complexes and promote the uniformity of the photoluminescence of V_{Si} arrays in 4H-SiC.

2. Computational methodology

First-principles calculations are carried out by the projector-augmented wave (PAW) method, as implemented in the Vienna *ab initio* simulation package (VASP).[37,38] The wave functions are expanded by using the plane waves up to a kinetic energy cutoff of 500 eV. The Perdew–Burke–Ernzerhof revised for solids (PBEsol) functional with the GGA exchange correlation is adopted for the structural relaxation.[39] The supercell size and atomic positions are fully relaxed until the total energy per cell and the force on each atom converge to less than 1×10^{-6} eV and 0.01 eV/Å, respectively. For accurate bandgap energy and defect level description, the hybrid density functional of Heyd, Scuseria, and Ernzerhof (HSE06), which mixes 25% of screened Hartree–Fock exchange to the PBE exchange functional, is employed during the calculation of electronic properties.[38] H impurities are modeled in 128-atom 4H-SiC supercells. For Brillouin zone integration, the Monkhorst–Pack scheme with a Γ-centered $2\times2\times2$ special k-points mesh is used.[39] Defect formation energies of H are calculated by the well-established mixed k-point scheme.[42,43]

3. Results and discussion

3.1. Configurations

As shown in Fig. 1, the configurations of H in 4H-SiC include substitutional H at C sites (H_C), substitutional H at Si sites (H_{Si}), as well as interstitial H at the bonding center of Si–C (H_i^{bc}), interstitial H at the tetrahedral center of Si (H_i^{Si-te}) and C (H_i^{C-te}), and interstitial H at the octahedral center (H_i^{oc}). We note that the nonequivalent lattice sites of Si and C in 4H-SiC include the quasihexagonal (h) and quasicubic (k) sublattice sites. The difference of the formation energy of H_C (H_{Si}) at the k-site and h-site is negligible. So, we just construct H_C and H_{Si} at the k-site as a representative.

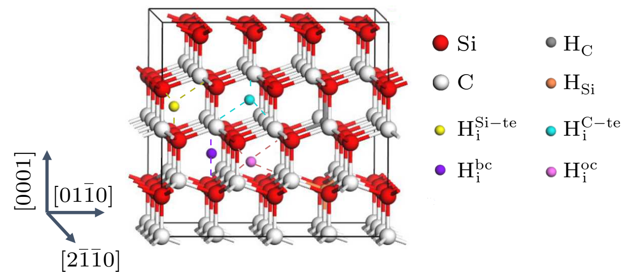

Fig. 1. Relaxed structures of H in 4H-SiC. H_C, H_{Si}, H_i^{bc}, H_i^{Si-te}, H_i^{C-te}, and H_i^{oc} are denoted by gray, orange, purple, yellow, cyan, and pink balls, respectively. Si and C atoms are denoted by red and white balls, respectively.

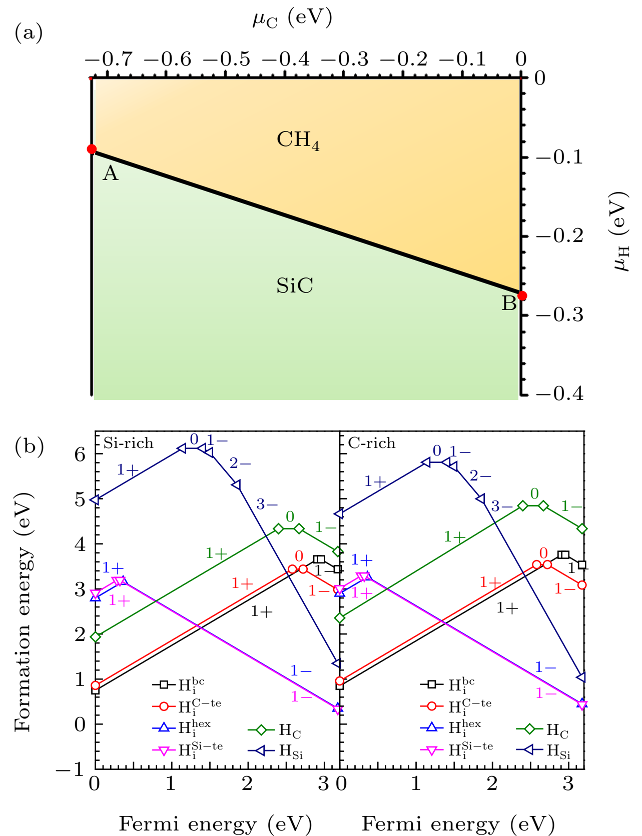

Fig. 2. (a) Accessible range of chemical potentials (green area) for equilibrium incorporation of H in 4H-SiC, (b) calculated formation energies of H in 4H-SiC.

During the incorporation of H in 4H-SiC, thermodynamic equilibrium growth conditions require a series of limitations

on the achievable values for chemical potentials of the constituents (μ_i). Firstly, the values of $\mu_{\rm Si}$ and $\mu_{\rm C}$ are limited to maintain the stable 4H-SiC:

$$\mu_{\rm Si} + \mu_{\rm C} = \Delta H_{\rm f}(\text{4H–SiC}), \tag{1}$$

where $\Delta H_{\rm f}$(4H–SiC) is the formation energy of 4H-SiC. The calculated $\Delta H_{\rm f}$(4H–SiC) is -0.73 eV for formula which is in good agreement with experiments (-0.68 eV, -0.75 eV or -0.771 eV for formula[44]). Secondly, to avoid the precipitation of elemental host phases and the elemental H, the values of μ_i are limited by

$$\mu_{\rm Si} \le 0, \quad \mu_{\rm C} \le 0, \quad \mu_{\rm H} \le 0. \tag{2}$$

Finally, CH_4(g) has the lowest formation energy. Due to this, the formation of CH_4(g) should be avoided. The values of μ_i are limited by

$$\mu_{\rm C} + 4\mu_{\rm H} \le \Delta H_{\rm f}({\rm CH_4}) = -0.79, \tag{3}$$

where $\Delta H_{\rm f}(\rm CH_4)$ are formation energy of CH_4. By solving Eqs. (1)–(3), we can get the accessible range for values of the chemical potentials, as shown by the green regions in Fig. 2(a). We then take the Si-rich limit [point A in Fig. 2(a)] and the C-rich limit growth condition [point B in Fig. 2(a)] to calculate the formation energies of H in 4H-SiC.

As shown in Fig. 2(b), the formation energies of H interstitials are lower than those of substitutional H, as a result of the steric effect. $\rm H_i^{bc}$ and $\rm H_i^{Si-te}$ dominate the configurations of H in p-type and n-type 4H-SiC, respectively. For $\rm H_i^{bc}$, the neutral charge state is never stable, because of the Column effect between $\rm H_i^{bc}$ and its neighboring atoms. When the Fermi energy increases up to 2.80 eV, the charge state of $\rm H_i^{bc}$ charges directly from 1+ to 1−. $\rm H_i^{Si-te}$ also acts as a negative-*U* center, with the charge transition from 1+ to 1− occuring at 0.34 eV. We find that $\rm H_i^{bc}$ and $\rm H_i^{Si-te}$ introduce deep defect states under the conduction band minimum (CBM) and above the valance band maximum (VBM) of 4H-SiC, respectively. These defect states are both occupied by one electron, indicating H may capture carriers and affect the performance of 4H-SiC (Fig. 3). More importantly, electron transfer between H and other defects (*e.g.*, dopants and intrinsic defects) would give rise to H-passivation of these defects. Unintentional H passivation of dopants would change the electronic properties of semiconductors.[45] During the processing of semiconductor devices, passivation of H is also frequently adopted to suppress the negative effect of intrinsic defects.[46,47] Therefore, we evaluate the effect of H passivation of the common n-type dopant (N), p-type dopant (Al), and intrinsic defects ($\rm V_C$ and $\rm V_{Si}$) on the performance of 4H-SiC. Because the growth conditions for both the single crystal growth and homoepitaxy of 4H-SiC are Si-rich, and the growth condition does not significantly affect the formation energies of H in 4H-SiC [Fig. 1(b)], we investigate the effect of H on the electronic properties of 4H-SiC under the Si-rich limit.

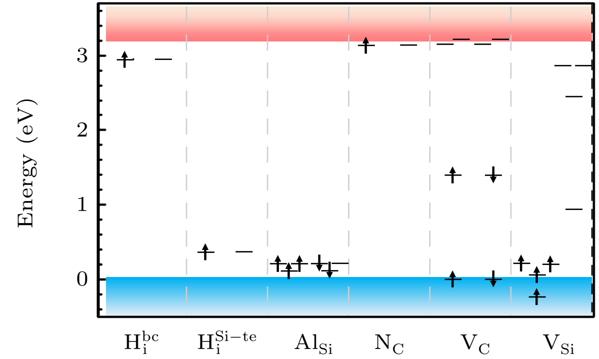

Fig. 3. Single-electron levels of dominant H interstitials, dopants, and intrinsic defects in 4H-SiC. Pink and blue regions indicate the CB and the VB of 4H-SiC, respectively. The up and down arrows denote the occupied spin-states of electrons on the defect levels.

3.2. H in n-type 4H-SiC

The high dopability of N in 4H-SiC endows 4H-SiC a great success in high power electronics. For n-type 4H-SiC substrates, the resistivity in the order of 10^{-3} Ω·cm is highly desired to guarantee its conductivity. During the homoepitaxy of n-type 4H-SiC layers, the electron concentration of the n-type buffer layer should be as high as possible to guarantee the conversion efficiency of basal plane dislocations.[48,49] However, the doping efficiency of N still lags behind the demand of ideal power-device applications. In this section, we evaluate the effects of compensation (or passivation) of H on the electronic properties of n-type 4H-SiC.

As shown in Fig. 4(a), the +/0 transition energy level of N locates at 0.06 eV below the CBM of 4H-SiC, which agrees well with theoretical and experimental results.[1] We find that the Fermi energy of 4H-SiC tends to shift toward its CBM as the concentration of N increases [Fig. 5(a)]. For n-type 4H-SiC, the formation energy of the negatively charged $\rm H_i^{Si-te}$ becomes lower than that of $\rm N_C^+$, which leads to the compensation of negatively charged $\rm H_i^{Si-te}$ on the n-type doping of 4H-SiC. The Fermi energy would be pinned in the mid-gap of 4H-SiC when the concentration of unintentional induced H becomes nonnegligible.

We also evaluate whether H can passivate $\rm N_C$ in n-type 4H-SiC. As shown in Fig. 4(a), the formation energy of $\rm N_C$–H complex is much higher than those of individual $\rm N_C$ and $\rm H_i$, indicating that the passivation of H at $\rm N_C$ is unlikely to happen under thermodynamic equilibrium growth conditions. We also calculate the binding energy of the $\rm N_C$–H complex [$E_{\rm b}$ ($\rm N_C$–H)] by $E_{\rm b}(\rm N_C\text{–}H) = \Delta H_f(N_C) + \Delta H_f(H_i) - \Delta H_f(N_C\text{–}H)$. The negative value of $E_{\rm b}$ ($\rm N_C$–H) throughout the bandgap of 4H-SiC indicates that the $\rm N_C$–H complex is not stable against decomposition. This indicates that the compensation of $\rm H_i^{Si-te}$ exerts more significant effect on the properties of n-type 4H-SiC. Reducing the concentration of unintentional

induced H is highly desired to improve the n-type doping and reduce the resistivity of highly N-doped 4H-SiC.

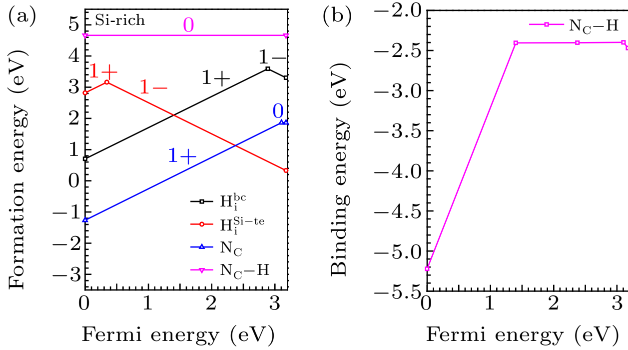

Fig. 4. (a) Calculated formation energies of N_C, N_C–H, H_i^{Si-te}, and H_i^{bc} in 4H-SiC, (b) calculated binding energy of the N_C–H complex in 4H-SiC.

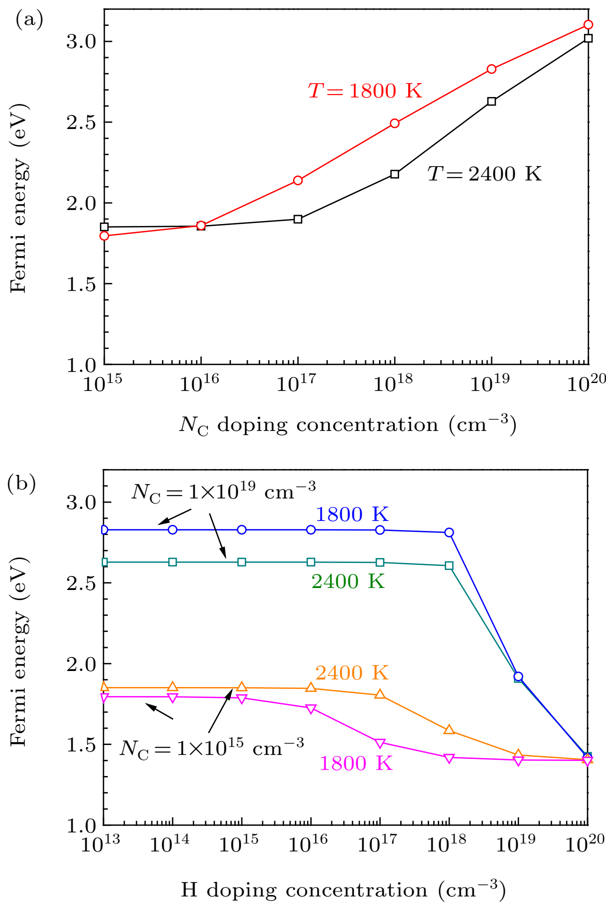

Fig. 5. (a) Fermi energies of 4H-SiC grown at 1800 K and 2400 K as functions of the concentration of N, (b) Fermi energies of 4H-SiC grown at 1800 K and 2400 K as functions of the concentration of H under different background concentrations of N.

The typical temperatures of PVT growth and CVD homoepitaxy of 4H-SiC are 2400 K and 1800 K, respectively.[1] Therefore, we investigate the effects of H on the Fermi energy and carrier concentration of n-type 4H-SiC at the temperatures of 2400 K and 1800 K. As shown in Fig. 5(a), the Fermi energy of 4H-SiC grown under 1800 K is slightly higher than that of 4H-SiC grown under 2400 K, due to lower concentration of thermally excited electrons at lower temperatures. This indicates that the hole concentration of 4H-SiC grown under 1800 K is higher than that of 4H-SiC grown under 2400 K. When the concentration of N increases from 10^{15} cm^{-3} to 10^{16} cm^{-3}, the increase of Fermi energy of 4H-SiC grown under 1800 K is more significant than that for 4H-SiC grown under 2400 K. For 4H-SiC grown under lower temperatures, the hole concentration is lower, which means that it is easier to tune the Fermi energy of 4H-SiC by increasing the concentration of N. When the concentration of N increases to 10^{19} cm^{-3}, the Fermi energy of 4H-SiC grown under 1800 K and 2400 K increase to 2.6 eV and 2.8 eV, respectively. Taking 4H-SiC with the N-doping concentration of 10^{19} cm^{-3} as an example, we find that the effect of unintentional incorporated H exerts negligible effect on the n-type doping of 4H-SiC. Only when the concentration of H exceeds 10^{18} cm^{-3}, the Fermi energy of 4H-SiC begins to decrease [Fig. 5(b)]. When the concentration of H increases to 10^{20} cm^{-3}, the Fermi energy of 4H-SiC is pinned at 1.4 eV due to the self-compensation of H [Fig. 5(b)].

3.3. H in p-type 4H-SiC

The p-type 4H-SiC substrates are of great importance to the development of n-channel bipolar devices based on 4H-SiC, which hold great promise for ultra-high voltage (> 10 kV) applications.[50–53] Al owns the lowest ionization energy among all group-III elements in 4H-SiC, which makes it as the most popular p-type dopant in 4H-SiC. It is well known that the lowest defect configuration of Al is Al_{Si}. Therefore, we investigate the interaction of H with Al_{Si} in this section, the effect of dominant intrinsic defect of V_C is also taken into consideration.

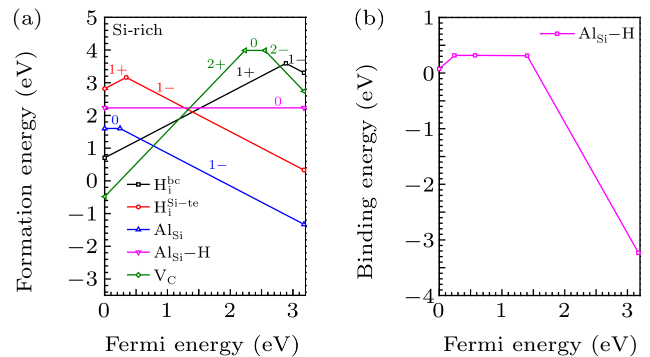

Fig. 6. (a) Calculated formation energies of Al_{Si}, Al_{Si}–H, V_C, H_i^{Si-te}, and H_i^{bc} in 4H-SiC, (b) calculated binding energy of the Al_{Si}–H complex in 4H-SiC.

As shown in Fig. 6(a), the formation energy of V_C is much lower than that of H_i^{bc}, indicating the compensation center for p-type 4H-SiC is V_C, rather than H. Because the formation energy of Al_{Si}–H complex is higher than those of individual Al_{Si} and H_i^{C-Si}, H is unlikely to passivate Al_{Si} under thermal equilibrium growth conditions. This ensures the effective doping of Al_{Si} in 4H-SiC. Nonequilibrium approaches, such as H ion implantation or radiation may passivate Al_{Si} by H. The

passivation can be easily eliminated by thermal annealing, because of the low binding energy of Al_{Si}–H in p-type 4H-SiC [Fig. 6(b)].

3.4. H passivation of V_C

When 4H-SiC is applied in power electronics, V_C has been identified as the carrier-lifetime killer in bipolar devices based on 4H-SiC.[26–30] By trapping carriers in its deep defect level, V_C severely reduces the carrier lifetime and thus the blocking voltage of the 4H-SiC drift layer.[53] Various approaches such as carbon ion implantation followed by thermal annealing, thermal oxidation, and annealing with a carbon cap, have been proposed to eliminate V_C in 4H-SiC.[54–57] For the sake of reducing processing complexity and cost, we evaluate whether H can passivate the defect states of V_C.

As shown in Fig. 2, passivating V_C by 4 H atoms can effectively eliminate the defect level of V_C and potentially improve the carrier lifetime in 4H-SiC. Therefore, we calculate the defect formation energies of V_C–nH (n = 1–4) to verify whether H is capable of passivating V_C and enhance the carrier lifetime of 4H-SiC. As shown in Fig. 7(a), the defect formation energies of V_C–nH (n = 1–4) are all larger than that of pure V_C. Although the deep defect states of V_C–4H disappears in 4H-SiC, V_C–4H has the highest formation energy among all V_C–nH (n = 1–4) complexes. This indicates that equilibrium incorporation of H cannot eliminate the defect states of V_C. Nonequilibrium approaches, such as H ion implantation or irradiation may capable of passivating V_C by H. The binding energy of V_C–nH (n = 1–4) complexes are calculated by $E_b(V_C\text{–}n\text{H}) = \Delta H_f(V_C) + n\Delta H_f(H_i) - \Delta H_f(V_C - n\text{H})$. As shown in Fig. 7(b), when the Fermi energy of 4H-SiC is in the range from 0.44 eV to 3.03 eV, the binding energies of V_C–4H is positive, indicating the V_C–4H complex is stable against decomposition. This means that nonequilibrium passivation of V_C by H is effective to eliminate the defect states of V_C and thus enhance the carrier lifetime of moderately doped 4H-SiC with Fermi energy ranging from 0.44 eV to 3.03 eV.

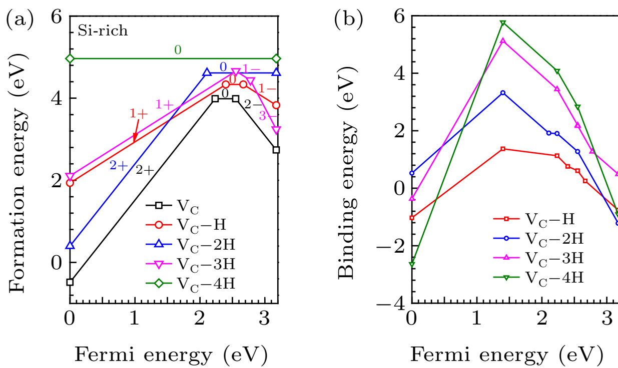

Fig. 7. (a) Formation energies and (b) binding energies of V_C–nH (n = 1–4) complexes in 4H-SiC.

3.5. H passivation of V_{Si}

Since 2015, V_{Si} has been manipulated as isolated spin qubits for quantum computing.[58–63] Isolated V_{Si} color centers were created by laser writing, ion implantation or electron irradiation.[58] It was found that both the photoluminescence (PL) wavelength and intensity were not uniform throughout the V_{Si} array of 4H-SiC.[64,65] In this section, we evaluate the possible reason of H passivation on the nonuniform PL emission of V_{Si} arrays in 4H-SiC.

As shown in Fig. 8(a), $(V_{Si}\text{–}4\text{H})^0$ and $(V_{Si}\text{–}3\text{H})^-$ complexes have the lowest formation energies in p-type and n-type 4H-SiC, respectively. This indicates that H would passivate V_{Si} and change the optical properties of V_{Si} defects in 4H-SiC. The occupied defect state of V_{Si} lies in 0.72 eV above the VBM of 4H-SiC, which agrees well with experimental and theoretical results.[40,41] When V_{Si} is passivated by 4 H atoms, the defect state disappears from the bandgap of 4H-SiC. This gives rise to the disappear of PL emission of V_{Si}. When V_{Si} is passivated by 3 H atoms, the defect states shift to 0.70 eV above the VBM of 4H-SiC. This results in the change of PL emission wavelength and intensity for the V_{Si} arrays in 4H-SiC.[66]

We also calculate the binding energies of V_{Si}–nH (n = 1–4) complexes by $E_b(V_{Si}\text{–}n\text{H}) = \Delta H_f(V_{Si}) + n\Delta H_f(H_i) - \Delta H_f(V_{Si}\text{–}n\text{H})$. As shown in Fig. 8(b), the binding energies of V_{Si}–nH (n = 1–4) complexes are all positive. This indicates that once the complexes are formed, V_{Si}–nH (n = 1–4) complexes are stable against decomposition. Because of the low binding energies of V_{Si}–nH (n = 1–4) complexes, thermal annealing is needed to decompose V_{Si}–nH (n = 1–4) complexes, and promote the uniformity for the PL properties of V_{Si} array in 4H-SiC.

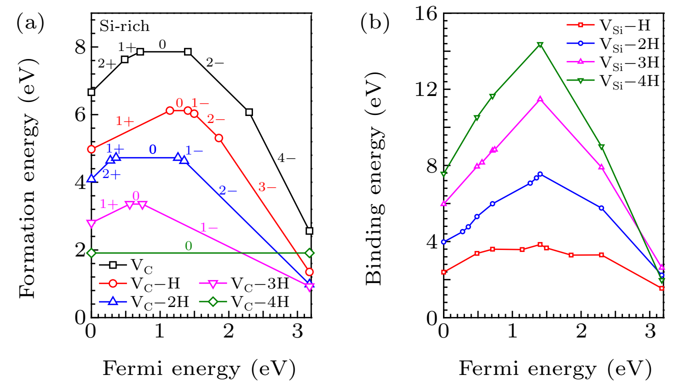

Fig. 8. (a) Formation energies and (b) binding energies of V_{Si}–nH (n = 1–4) complexes in 4H-SiC.

4. Conclusion

In conclusion, we have systematically investigated the role of H in the electronic properties of 4H-SiC. We have

found that H_i^{bc} and H_i^{Si-te} dominate the defect configurations of H in p-type and n-type 4H-SiC, respectively. In n-type 4H-SiC, the compensation of H_i^{Si-te} is found to pin the Fermi energy and hinder the increase of electron concentration in highly N-doped 4H-SiC. Reducing the concentration of unintentional incorporated H is critical to further reduce the resistivity of n-type 4H-SiC. For p-type 4H-SiC doped by Al, we find that the compensation of H on the p-type doping of 4H-SiC is negligible compared to that of V_C. Nonequilibrium H-incorporation approaches may passivate Al_{Si} by H, and degrades the performance of p-type 4H-SiC. The degradation can be easily eliminated by thermal annealing, because of the low binding energy of the Al_{Si}–H complex in p-type 4H-SiC. We find that nonequilibrium passivation of V_C by H is effective to eliminate the defect states of V_C and thus enhance the carrier lifetime of moderately doped 4H-SiC. Regarding the quantum-qubit applications of 4H-SiC, we find that H can readily passivate the silicon vacancy (V_{Si}) during the creation of V_{Si} centers. The V_{Si}–nH ($n = 1$–4) complexes are stable against decomposition once they are formed. Thermal annealing is needed to decompose the resulting V_{Si}–nH ($n = 1$–4) complexes and promote the uniformity of the photoluminescence of V_{Si} arrays in 4H-SiC. The current work may inspire the further development of the impurity engineering of H in 4H-SiC.

Acknowledgments

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