Point Defects in Monolayer *h*-AIN as Candidates for Single-Photon Emission

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ABSTRACT: A single-photon emission (SPE) system based on a solid state is one of the fundamental branches in quantum information and communication technologies. The traditional bulk semiconductors suffered limitations of difficult photon extraction and long radiative lifetime. Two-dimensional (2D) semiconductors with an entire open structure and low dielectric screening can overcome these shortcomings. In this work, we focus on monolayer *h*-AlN due to its wide band gap and the successful achievement of SPE compared to its bulk counterpart. We systematically investigate the properties of point defects, including vacancies, antisites, and impurities, in monolayer *h*-AlN by employing hybrid density functional theory calculations. The -1 charged Al vacancy (V_{AI}⁻) and +1 charged nitrogen antisite (N_{AI}⁺) are predicted to achieve SPE with the zero-phonon lines of 0.77 and 1.40 eV, respectively. Moreover, the charged point-



defect complex $C_{Al}V_N^+$, which is composed of vacancies and carbon substitutions, also can be used for SPE. Our results extend the avenue for realizing SPE in 2D semiconductors.

KEYWORDS: quantum communication, single-photon emission (SPE), monolayer h-AlN, point defects, optoelectronic properties, hybrid density functional theory

INTRODUCTION

Rapid progress has been made in quantum information technologies in the last two decades, which depends on the development of light sources capable of controllable photons. Recently, single-photon emission (SPE) based on a solid-state platform has been shown to be the most necessary branch in this field.¹⁻⁵ To achieve SPE, following two requirements should be satisfied: (1) there should be distinctive two-level quantum states, a ground state and an excited state, in the same spin channel to meet spin conservation during light stimulation and emission; (2) these states should be energetically insulated from the bulk states to minimize the external interference.⁶ The defect levels in a wide-gap semiconductor can meet these criteria. First, the defect state is usually quite localized and isolated. Second, the wide band gap supports enough space to modify the emitting photons. In the past, SPE has been successfully realized in many solid-state systems, such as defect complexes in diamonds (negative nitrogen substitution around vacancy, NV^- , and silicon substitution around vacancy, SiV,⁷⁻¹⁰ point defects in different polytypes of silicon carbide,^{6,11} and quantum dots.^{12,13} III-Nitrides were appealing to fabricate SPE devices due to their promising properties¹⁴ and mature development of related industries.^{15,16} Satoshi and co-workers realized SPE based on gallium nitride (GaN) quantum dots at 200 K.¹⁷ Amanuel demonstrated that the defects induced by the

stacking fault act were the emission center in GaN.¹⁸ Aluminum nitride (AlN), possessing a wider band gap¹⁹ than GaN, therefore, may realize high-energy SPE to satisfy the usage of free-space quantum connection.²⁰ The feasibility of SPE based on V_N-related centers in AlN was predicted by firstprinciples calculations, and the properties were similar to those of NV⁻ centers in diamond.²¹ Xue and co-authors experimentally realized SPE that ranged from visible to nearinfrared regions by utilizing defect complexes $(N_{Al}V_{N}%)$ and V_{Al}V_N) hosted in AlN.²² Yet, these systems suffered shortcomings, such as difficult photon extraction in bulk materials due to high reflection index and long radiative lifetime.^{4,7,23} Even for the most studied NV⁻, SiV center, etc., in diamond, the brightness of the single-photon source was still low owing to the low quantum efficiency; however, the NV⁻ showed spindependent fluorescence and the SiV exhibited a very narrow emission bandwidth.⁹

The atomically thin two-dimensional (2D) materials exhibit natural advantages for SPE. The entire openness of the

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structure enabled all atoms to expose in light that will increase the efficiency of light absorption. The excellent light transparency almost eliminated the scattering in bulk materials, thus facilitating the extraction of emitted photons.⁴ The effective defect Bohr radii of 2D materials were reduced due to weakened dielectric screening,^{24,25} and as a result, the defect level became deeper and more localized, which is beneficial for the SPE. At the same time, the radiative rate²⁶ and oscillator strength²⁷ were enhanced with the reduction of dielectric constants in 2D materials by comparing with the bulk phase, which was helpful to improve the quantum efficiency of the optical transition between two-level states in 2D semiconductors. In fact, the wide-gap 2D semiconductor hexagonal boron nitride (h-BN) has attracted considerable attention for implementing quantum qubits,^{28,29} even though the microscopic origin of the SPE in h-BN is still being debated.^{21,30} The zero-phonon lines (ZPLs) in the range of 1.6-2.2 eV have been observed in h-BN after electron irradiation or annealing treatment.³¹ Notably, the ZPLs of the samples grown by lowpressure chemical vapor deposition were in the range of 2.10-2.18 eV.³² Moreover, the defects in 2D Mo- or W-based transition-metal dichalcogenides had already been theoretically predicted^{4,5} and experimentally achieved stable SPE.³³ Developing more materials is necessary to further improve the performance of SPE sources. 2D III-nitrides semiconductors show great potential to provide more options in this field not only due to the chemical comparability to h-BN but also due to the mature development of their bulk phases, such as deep UV light-emitting diodes,^{34,35} lasers diodes,^{36,37} and transistors.³⁸ Following theoretically confirming the good thermodynamic stability of hexagonal AlN (h-AlN),²⁴ Tsipas et al., Mansurov et al., and Malin et al. had experimentally realized *h*-AlN via molecular beam epitaxy.^{39–41} The defective monolayer *h*-AlN is highly expected to be a good host material for qubits due to its similarity to *h*-BN. In addition, the properties of AlN nanosheets,⁴² nanoribbons,^{43,44} hetero-junctions,⁴⁵ functionalization,^{46,47} and magnetic dopants^{48,49} for h-AlN were also reported. However, studies on the native point defects in h-AlN and their applications are still limited. Almeida et al.⁵⁰ explored the structural and electronic properties of native point defects and C/Si impurities, and López-Pérez et al.⁵¹ further considered charged vacancies.

Here, we revealed the stability and electronic properties of point defects in monolayer *h*-AlN and evaluated the potential to realize SPE by first-principles calculations based on hybrid density functional theory (DFT). Total nine defects, including vacancies (V_{Al} and V_N), antisites (Al_N and N_{Al}), impurities (C_N , C_{Al} , Mg_{Al} , Si_{Al} , and O_N), and their charged states were considered. The V_{Al} can be stable and host two-level quantum system. Moreover, the point defect complexes $C_{Al}V_N$ and C_NV_{Al} were also discussed.

METHODS

First-principles calculations based on DFT were performed for the electronic structure analysis. The band structure and defect formation energy were calculated with the Vienna Ab Initio Simulation Package (VASP),⁵² where the projector-augmented wave method was adopted. The geometry optimization was calculated with the Perdew–Burke–Ernzerhof (PBE)⁵³ functional within the generalized gradient approximation. A dispersion correction (DFT-D3) was used to incorporate the long-range vdW interaction.^{54,55} The plane-wave basis set with a kinetic energy of 500 eV was used. We used a $6 \times 6 \times 1$ supercell to study the properties of defects in *h*-AlN; the lattice constant along the *h*-AlN plane was about 18.94 Å, and the vacuum

layer was set to be 20 Å to separate the interactions between neighboring layers. We employed $2 \times 2 \times 1$ Monkhorst–Pack *k*-grids in self-consistent calculations (free energies were well converged) and 20 *k*-points between high symmetry points of the Brillouin zone in the band structure calculations; in addition, the force and total energy convergence thresholds were set to 0.01 eV/Å and 10^{-5} eV, respectively.

To assess the SPE property exactly, the geometry and electronic properties were redetermined by the Heyd–Scuseria–Ernzerhof (HSE06) hybrid functional,⁵⁶ where the ratio of exact exchange was set to be 0.33. The HSE06-related calculations were carried out by GPU-accelerated PWmat software,⁵⁷ which was much faster than VASP. The excited-state geometry was calculated by the constrained DFT method, in which the occupation of the electrons was fixed during relaxation. The ZPL emission was obtained by the adiabatic energy difference between ground and excited states.

In this work, the formation energy (E_{form}) of the point defects *D* at charge states *q* is defined by the following formula (eq 1):^{58,59}

$$E_{\text{form}}[D^{q}] = E_{\text{tot}}[D^{q}] - E_{\text{tot}}[\text{host}] - \sum_{\alpha} n_{\alpha}\mu_{\alpha} + q(E_{\text{VBM}} + E_{\text{F}} + \Delta V)$$
(1)

where $E_{\rm tot}[D^q]$ and $E_{\rm tot}[{\rm host}]$ are the total energies of the supercell with point defects at the *q*-charged state and host monolayer *h*-AlN, respectively, n_{α} is the number of defect atoms from the monolayer, and μ_{α} is the chemical potential of the species α in their stable elementary phase. The atomic chemical potentials for their most stable elementary phases were used in the formation energy calculations: C in the graphite phase, Si in the diamond phase, Mg in the hcp phase, Al in the fcc phase, and N and O in the gas phase. $E_{\rm VBM}$ is the valence band maximum (VBM) energy of the perfect *h*-AlN, $E_{\rm F}$ is the Fermi energy level with respect to VBM, which was considered to be zero if it coincided with VBM, and ΔV is the electrostatic potential alignment correction. In this work, we have considered +2, +1, 0, -1, and -2 for native point defects, only +1 for n-type impurities, and only -1 for p-type impurities.

The calculated E_{form} was used to further evaluate the defect density n in h-AlN at a finite temperature T through the Arrhenius equation (eq 2):

$$n = n_0 \exp\left(\frac{-E_{\rm form}}{k_{\rm B}T}\right) \tag{2}$$

where n_0 is the areal atom density, E_{form} is the formation energy of the defect, and k_{B} is the Boltzmann constant. The defect ionization level $\varepsilon(q/q')$ corresponds to two defect stable charge states q/q'. The ionization level is defined as eq 3:

$$\varepsilon(q/q') = \frac{E_{\text{form}}[D^q] - E_{\text{form}}[D^{q'}]}{q' - q}$$
(3)

where $E_{\text{form}}[D^q]$ is the formation energy of the charged defect with the Fermi level set at the VBM. Here, the WLZ extrapolation method was employed to correct the error induced by artificial long-range Coulomb interactions along the vacuum direction in 2D systems.⁶⁰ For thinner 2D materials, the calculation of ionization energy of a defect with charge *q* was calculated according to eq 4:

IE
$$(S, L_z) = IE_0 + \frac{\alpha}{\sqrt{S}} + \frac{\beta}{S}L_z$$
 (4)

where IE(S, L_z) is the defect ionization energy $\varepsilon(q/q')$, IE₀ is the real size-independent, vacuum-height-independent transition level, α corresponds to the defect-specific Madelung constant, β corresponds to $q^2/24\varepsilon_0$, and S and L_z are the square of the supercell and vacuum height, respectively. In this work, the dimensions of the supercell were selected to be 20 and 40 Å for L_z with 6 × 6 and 8 × 8 supercells for $L_x \times L_y$, respectively, for each structure.

The absorption spectrum was calculated with the random phase approximation (RPA) method, which was based on direct Fermi's golden rule (eq 5):

$$\varepsilon_{2}(\omega) = \sum_{i \in VB, j \in CB} \left| \left\langle \psi_{i} \middle| \frac{\partial H}{\partial k} \middle| \psi_{j} \right\rangle \right|^{2} \delta(E_{j} - E_{i} - \omega)$$
(5)

where $\partial H/\partial k$ is the momentum operator and E_i is the eigenvalue of state $|\psi_i\rangle$. The radiative lifetime was calculated by Fermi's golden rule, and the concrete form of the radiative rate was given by eq. 6:

$$W_{\rm rad}(\omega) = \frac{\omega^3 n \left| \mu_{ij} \right|^2}{3\pi \varepsilon_0 \hbar c^3} \tag{6}$$

where ω is the frequency of the emitted photon, n is the index of refraction, μ_{ij} is the transition dipole moment, ε_0 is the vacuum permittivity, \hbar is the reduced Planck constant, and c is the vacuum speed of light. According to the relationship $|\mu_{ij}|^2 = |\langle \psi_i | r | \psi_j \rangle|^2 = |\langle \psi_i | r | \psi_j \rangle|^2 = |\langle \psi_i | r | \psi_j \rangle|^2 \times (\hbar^2 / \Delta E_{ij}^2 m_e)$, the radiative rate $W_{\rm rad}(\omega)$ and radiative lifetime $\tau_{\rm rad} = 1/W_{\rm rad}(\omega)$ can be obtained.

RESULTS AND DISCUSSION

The lattice parameters of wurtzite AlN obtained through our method (a = 3.11 Å and c = 4.98 Å) agree with the reported experimental⁶¹ and theoretical results,⁶² indicating the good reliability of our method. The crystal structure of monolayer *h*-AlN is shown in Figure 1a; the symmetry of the crystal has a



Figure 1. (a) Optimized structures of monolayer h-AlN; purple and blue are Al and N atoms, respectively. (b) Band structure of the prefect h-AlN obtained at PBE (black solid lines) and HSE06 (blue dashed lines) levels. Insets are the spatial distributions of the squared wave function for CBM and VBM, respectively.

space group P6/m2 and point group D_{3h} . Each Al (or N) atom is threefold coordinated to N (or Al) atoms through double bonds composed of one sp² bond and one π bond, resulting in a honeycomb pattern. The optimized lattice parameter and Al-N bond lengths are calculated to be 3.12 and 1.80 Å, respectively. The electronic band structure for monolayer h-AlN is plotted in Figure 1b. An indirect band gap of 4.43 eV is observed, instead of the 6.20 eV direct band gap (bulk wurtzite AlN). The reduction of the band gap from the bulk to the monolayer is due to the chemical bond change from sp³ to sp² hybridization, as in the case of silicon and silicene.⁶³ By comparing with the band structure of h-BN,56 that of h-AlN exhibits similar dispersion near the VBM but different dispersion near conduction band minimum (CBM). The nearly free electron states near CBM (at k point) for h-BN are not observed in h-AlN. This should be attributed to the different origination of band edges. As shown in the insets of Figure 1b, the VBM is derived from π -bonding states of N $2p_z$ for both *h*-BN and *h*-AlN and the CBM is derived from π^* and σ^* antibonding states hybridized by N 2s and Al 3p_z orbitals for *h*-AlN but solely from N $2p_z$ for *h*-BN. The properties of the perfect monolayer h-AlN are well reproduced, and the results are reported in previous works.^{47,51}

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Here, we discuss the formation energy (E_{form}) of the native point defects and impurities. For vacancies, the E_{form} values of V_{Al} are 10.60 and 8.93 eV under N-poor and N-rich conditions, respectively, implying the pretty low density of V_{Al} in materials, as shown in Table 1 and Figure 2, even though the

Table 1. Formation Energy $(E_{\rm form})$ of Native Point Defects in the Neutral State, in eV

defects	N-poor	N-rich
V _{Al}	10.60	8.93
V_N	3.05	4.72
Al _N	4.02	7.35
N _{Al}	9.30	5.97
C _{Al}	4.71	3.05
C _N	2.94	4.60
Mg _{Al}	3.37	1.70
Si _{Al}	1.40	-0.27
O _N	1.75	3.51

temperature is as high as around 2000 K. In contrast, the E_{form} values of V_N are 3.05 and 4.72 eV, which are much lower than those for V_{Al} . Therefore, the density of V_N can reach 10^4 and 10^8 m^{-2} under N-rich and N-poor conditions, respectively. We also consider charged vacancies. The q = +1 and q = -1charged V_{Al} can be stable, while the Fermi-level can be lower than 0.07 eV above VBM and higher than 1.33 eV above VBM, as shown in Figure 2. The V_N can be stable at q = +2, +1, and -1 charged states with the thermodynamic transition levels (ε) of 0.19, 3.27, and 3.80 eV above VBM, respectively. For antisites, the space for the Al site is much larger than that for the N site because the Al atom possesses a larger atomic radius. Therefore, the Al atom is squeezed out of the plane for Al_N , but the N_{Al} structure generally remains flat. In the neutral state, Al_N is more easily formed than N_{Al} under N-poor conditions but not under N-rich conditions. The density can reach 10⁹ m^{-2} under N-poor conditions for Al_N and $10^4 m^{-2}$ under Nrich conditions for N_{Al} at 2000 K. Moreover, $Al_{\rm N}{}^{-},\,N_{Al}{}^{+},$ and N_{Al}^{-} are stable with ε values of 1.13, 2.76, and 3.92 eV, respectively. For impurities, the $E_{\rm form}$ values are generally smaller than those for vacancies and antisites. Notably, the E_{form} of Si_{Al} under N-rich conditions is negative (-0.27 eV), implying that h-SiN may be a stable structure. The q = -1states for p-type impurities are more stable than neutral states when the Fermi level is close to the CBM, and the q = +1states for n-type impurities are more stable than neutral states when the Fermi level is close to the VBM.

Next, we will discuss the electronic and magnetic properties of these defect structures. The vacancy is formed by removing an Al or N atom. After structural relaxations, the D_{3h} symmetry is lowered to C_s due to the out-of-plane displacement of atoms neighboring the vacancies. The absence of the Al or N atoms in monolayer *h*-AlN leaves dangling bonds (three sp² and three p_z) on three nearest-neighbor N or Al atoms. The molecular orbitals near the vacancies can be constructed by linear combinations of these states. As a result, localized symmetric (a') and higher-lying antisymmetric (a'') molecular orbitals are formed. The V_{Al} can accept at most three electrons according to the electron count, therefore indicating a triple acceptor character. In the neutral state, three holes appear in the highlying states of the spin-down channel, corresponding to a high-

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Figure 2. Formation energies (E_{form}) as a function of the Fermi level for native point defects in *h*-AlN under (a) N-rich and (b) N-poor conditions. The solid and dashed lines represent native defects and impurities, respectively. The Fermi level can vary from VBM (set as zero) to CBM (4.43 eV).



Figure 3. Spin-charge density of optimized structures of the considered (a) neutral and (b) stable charged native point defects in 2D AlN. The isosurfaces exhibit the spin-charge density for each structure; red and yellow represent spin-up and spin-down channels, respectively. (c) Thermodynamic transition levels for the considered point defects and impurities in h-AlN.

spin paramagnetic state; this can be also found in the calculated band structures shown in Figure S1. The calculated magnetic moment is 3 $\mu_{\rm B}$. As the spin-charge density shown in Figure 3a, the magnetic moments are localized around the dangling N atoms most neighboring the vacancy. In the V_{Al}^{+} and V_{Al}^{-} states, an electron is extracted from (or added to) one a'' state, and as a result, the magnetic moment reduces to 2 $\mu_{\rm B}$ as shown in Figure 3b. The $\varepsilon(+/0)$ and $\varepsilon(0/-)$ for V_{Al} are 0.07 and 1.33 eV, respectively (Figure 3c), which are shallower than those in bulk AlN.⁵⁷ On the other hand, the removal of a N atom from the h-AlN lattice leaves three Al $3sp^2$ and three Al $3p_z$ dangling bonds, which combine with low-lying a' states and higher-lying a'' states. Electron counting indicates that the three electrons occupy the lowest a' defect orbitals. Therefore, it is a doublet in the spin-down channel. The magnetic moment is 1 $\mu_{\rm B}$ (as shown in Figure 3a), corresponding to a low-spin state as compared to the V_{Al} . The spin moment is located around the p_z orbital of dangling Al atoms. These results are comparable to those for *h*-BN.⁶⁴ The $\varepsilon(2+/+)$, $\varepsilon(+/$ 0), and $\varepsilon(0/-)$ for V_N are 0.19, 3.27, and 3.80 eV, respectively, as shown in Figure 3c, which are all deep defect states. The Al atom is substituted by a N atom to form an antisite defect NAI. In this case, the gap states are formed by the localized homoelemental bonds (N-N bond). Nitrogen contains two more valence electrons than aluminum, so in the neutral state, N_{Al} introduces two extra electrons, resulting in a nonmagnetic ground state. As expected, the calculated total magnetic moment is zero. From the band structures shown in Figure S2,

we find that the spin-up and -down channels are degenerated, which are broken in the N_{Al}⁺ and N_{Al}⁻ states. The 1 $\mu_{\rm B}$ magnetic moment is revealed and localizes around the defect (Figure 3b). The $\varepsilon(+/0)$ and $\varepsilon(0/-)$ for N_{Al} are 2.76 and 3.92 eV, respectively (Figure 3c). Similar results are observed for the Al_N case.

Impurities are unavoidable since the environment of material epitaxy is quite complex and because of the demands for functionalization. The metal-organic sources introduce carbon impurities, the oxygen impurities probably originate from the substrates, and Mg and Si are always used to realize the p- and n-type doping. The sizes of selected impurities are comparable to or smaller than those of the original sites; thus, the $C_{3\nu}$ symmetries are observed for these structures. In neutral states, the ground states of $C_{N'}$, Mg_{AV} and O_N are paramagnetic with 1, 1, and 0.6 $\mu_{\rm B}$ magnetic moments, respectively, and the magnetism localizes around the impurities. Meanwhile, the C_{AI} and Si_{Al} are nonmagnetic because the unpaired electron is quite delocalized, as shown in Figure S2. Moreover, we consider the charged states for these impurities. The *p*-type impurities (C_N and Mg_{Al}) prefer accepting one more electron to form q = -1 states; in contrast, the n-type impurities (C_{AV}) Si_{Ab} and O_N) prefer forming q = +1 states. We find that the ionization levels in Mg-doped and Si-doped h-AlN are quite deep in the gap than those in the corresponding bulk AlN,^{65,66} as shown in Figure 3c, indicating that it is still challenging to realize high-efficiency p- and n-type doping.

We analyze the optical properties and evaluate the possibility to apply in SPE. After screening all native point defect and impurity structures, we just discuss the structures with twolevel states and the calculated density of states (Figure S3). To obtain the more accurate defect-level diagrams (DLDs) for selected structures (only the states in the band gap are plotted), we repeat the structural relaxations with the HSE06 level. The two-level quantum system is observed in the spindown channel for V_{Al} , V_{Al}^{-} , and N_{Al}^{+} . For V_{Al} , the occupied and unoccupied levels are isolated by 0.35 and 0.94 eV from the host states, respectively, which are large enough to prevent interference. However, the intensity of absorption is much weaker than intrinsic states; moreover, the calculated radiative lifetime $(\tau_{\rm rad})$ is as long as 2.71 \times 10⁶ ns. The DLD, absorption, and coordinate diagram are shown in Figure S4. We can find from Figure S4b that the intensity for defect absorption is about one-fifth of that for intrinsic absorption; the long radiative lifetime should be attributed to the symmetry induced forbidden of the corresponding wavefunctions. Thus, the efficiency of SPE for V_{Al} may be quite low. The DLD and absorption are shown in Figure S4. As discussed above, the V_{Al} can also be stable. The two-level quantum system is observed in the spin-down channel and isolated by 1.15 and 0.33 eV for occupied and unoccupied states, respectively, for V_{Al}, as shown in Figure 4a. We examine



Figure 4. (a) Defect-level diagrams. (b) Calculated absorption coefficient. (c) Configuration coordinate diagrams of V_{Al}^- and N_{Al}^+ . In (a), the two-level quantum states are marked by dashed ovals in *h*-AlN. Blue and red arrows represent spin-up and -down channels, respectively. Solid and hollow arrows indicate occupied and unoccupied states, respectively. The values of ZPL for each structure are also marked. In (b), the first peaks are marked by dashed circles. In (c), the red, blue, and green arrows represent vertical excitation, ZPL, and vertical emission lines, respectively.

the excitation process by computing the absorption coefficient based on the dielectric constant. As shown in Figure 4b, the first peak of V_{AI}^- is at 1.08 eV and the intensity is very strong, which results in a short τ_{rad} by 128 ns. Therefore, the two-level quantum systems in V_{AI}^- can be excited via illumination. The coordinate diagram shown in Figure 4c reveals the excitation and emission process. The *x*-axis coordinate for the coordinate diagram is defined as $Q = \sqrt{\sum_i m_i (\vec{x_i} - \vec{x'_i})^2}$, where m_i

represents the mass of the *i*th atom and \vec{x}_i and \vec{x}'_i represent the initial and final coordinates of the ith atom, respectively. The vertical absorption energy is 1.36 eV, and the ZPL is 0.77 eV for V_{Al}^- , indicating that the energy loss by phonon scattering is 0.59 eV. For N_{Al}^+ , the two-level quantum system also appears in the spin-down channel, the excitation energy is 2.72 eV, and the calculated ZPL is 1.40 eV, as shown in Figure 4a,b. The energy scattered by phonon vibration is 0.63 eV. The τ_{rad} is as short as 74 ns. However, the excitation state of N_{Al}^+ is too close to the CBM. For V_{N} , Al_N , and impurities, the two-level quantum system can be observed neither in the neutral nor in their charged states. Among the native point defects, only the V_{Al}^- and N_{Al}^+ are suitable to realize SPE.

The defect complex, such as NV⁻ in diamonds or C_BV_N in *h*-BN,¹⁶ can host the qubits. Here, we also investigate the electronic properties of the vacancies combining with carbon substitution. In the defect complex, the carbon atom substitutes one of the dangling Al (or N) atoms neighboring the N (or Al) vacancy to form $C_{Al}V_N$ and C_NV_{Al} . The calculated E_{form} values are 5.82 and 9.50 eV for $C_{Al}V_N$ and C_NV_{Al} , respectively. In the neutral state, only one defect level can be observed in the band gap, and another energy level has emerged in the bulk materials so that these structures cannot be used for SPE. Furthermore, we consider both q = -1 and q = +1 states for $C_{Al}V_N$ and C_NV_{Al} . As shown in Figure 5, $C_{Al}V_N^+$ possesses an isolated two-level quantum system in the spindown channel with an energy of 3.59 eV, a τ_{rad} of 429 ns, and a ZPL of 2.30 eV.



Figure 5. (a) Defect-level diagram, (b) calculated absorption coefficient, and (c) configuration coordinate diagram (c) for $C_{AI}V_{N}^{+}$.

CONCLUSIONS

In summary, we have investigated the electronic properties of the native point defects and impurities in h-AlN and the potential of SPE realization by hybrid DFT calculations. The densities of native point defects are pretty low due to the high formation energies, even though the temperature as high as 2000 K. The native point defect levels are generally shallower, but the typical p- and n-type doping is much deeper than that in bulk AlN. The native point defects induce localized defect states, but the impurities induce delocalized defect states. The $V_{Al}{}^-$ and $N_{Al}{}^+$ can host SPE with the excitation energies of 1.08 and 2.72 eV, respectively. The calculated ZPLs are 0.77 and 1.40 eV. On the other hand, the V_N , Al_N , and impurities are not active for SPE. In addition, we find that the charged point defect complex C_{Al}V_N⁺ also exhibits the great potential to achieve SPE with a ZPL of 2.30 eV. These results will broaden the avenue to study quantum information science and technology.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsami.1c09175.

The PBE-level band structures for all neutral defects, spin-charge densities for the impurities, HSE06-level DOS for two-level systems, and defect-level diagram and calculated absorption coefficient for $V_{\rm Al}$ (PDF)

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Author Contributions

D.L., X.S., and Z.S. conceived the idea and provided advice on this work. H.Z., Z.Q., and Z.S. carried out the calculations and wrote the manuscript. Y.J., K.J., Y.C., S.Z., and X.S. provided very useful comments. All authors participated in the discussions on the results.

Notes

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