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1 **Comparative studies of interatomic potentials for modeling point defects in wurtzite**
2 **GaN**

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11 In this paper, a new version of the Stillinger-Weber (SW) potential for wurtzite
 12 GaN is presented, by which we systematically explore the structural and thermo-
 13 dynamical properties of native point defects and their complexes. In parallel, the
 14 semi-empirical Modified Embedded-Atom Method (MEAM) potential is selected for
 15 comparison. The SW and MEAM potentials are assessed by the reproduction of the
 16 fundamental properties of wurtzite GaN and by the ability to describe the inversion
 17 domain boundaries and the wurtzite-rocksalt phase transition. Then the structural
 18 search of native point defects and their complexes in GaN is implemented using both
 19 SW and MEAM potentials with the benchmark of Density Functional Theory (DFT)
 20 calculations. Besides vacancies and antisites, four N and five Ga interstitials are
 21 confirmed through the refining of DFT calculations, among which two N split in-
 22 terstitials $N^+-N\langle 2\bar{1}\bar{1}0 \rangle$, $N^+-Ga\langle 01\bar{1}0 \rangle$, and two Ga split interstitials $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -g,
 23 $Ga^+-N\langle 01\bar{1}0 \rangle$ are observed for the first time. The SW potential correctly predicts the
 24 octahedral occupation Ga_{Oct} to be the most stable Ga interstitial, while the MEAM
 25 potential predicts the ground state of $N^+-N\langle 01\bar{1}0 \rangle$ split interstitial ($N^+-N\langle 01\bar{1}0 \rangle$ -g) as
 26 the most stable N interstitial. But neither of two potentials could generate simultane-
 27 ously the most stable configurations of N and Ga interstitials. The investigations of
 28 point defect complexes reveal that N octahedral Frenkel ($Frenkel_{Oct}(N)$) and paired
 29 antisite ($N_{Ga}Ga_N$) defects are unstable and convert into the $V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g con-
 30 figurations with the different separations between V_N and $N^+-N\langle 01\bar{1}0 \rangle$ -g point defects
 31 based on the DFT calculations. The formation energies calculated by the DFT and
 32 SW potential demonstrate that Schottky, Ga octahedral Frenkel ($Frenkel_{Oct}(Ga)$)
 33 and $V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g point defect complexes are energetically feasible, and they
 34 should not dissociate into two isolated point defects. In contrast, the MEAM poten-
 35 tial predicts the dissociation to be exothermic for Schottky and $V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g.
 36 Overall, the structural features concerned with N-N or Ga-Ga bonds relaxed by the
 37 SW potential are more consistent with DFT calculations than the MEAM counter-
 38 part.

39 Keywords: GaN, Point defects, empirical potentials, First-principles calculation,
 40 Molecular statics, Molecular dynamics

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41 I. INTRODUCTION

42 GaN and related alloys attract scientific and industrial interests due to their unique
43 properties¹. They have direct band gap, high melting point, high thermal conductivity,
44 high breakdown field, high electronic mobility and high saturation electron drift velocity.
45 Belong to the $P6_3mc$ space group they also inherently possess spontaneous and piezoelectric
46 polarization. In GaN the strong bonding between cation and anion atoms gives it an intrinsic
47 thermal stability and high radiation resistance with the displacement threshold energy up
48 to 19 eV for Ga atoms². These merits make GaN-based alloys suitable for high power, high
49 temperature and high frequency electronic devices to operate in harsh environments such as
50 power supplies, automotive industries, aerospace communications and nuclear reactors³⁻⁶.
51 However, the performance of GaN-based devices is usually restricted below the theoretical
52 values. They suffer the permanent and/or reversible degradations due to the structural
53 defects, which mainly consist of point defects, threading dislocations and grain boundaries
54 *etc.*^{7,8}.

55 The point defects such as vacancies, antisites and interstitials could be generated
56 during non-stoichiometric growth, thermal excitation⁹, ion implantation¹⁰ or irradiation
57 damage^{11,12}, *etc.*. These native point defects and their complexes strongly influence the
58 electrical and optical properties of materials. They act as traps and recombination centers
59 for carriers^{7,13} thus to deteriorate the efficiency of devices¹⁴, and they may also impede
60 *p*-type doping in GaN by inducing compensation¹⁵. The point defects critically control
61 the diffusion dynamics in ion implantation, thermal annealing and recovery of irradiated
62 damage¹⁶. On the other hand, they could be purposefully created to improve the perfor-
63 mance of GaN-based devices. For instance, Ga interstitials generated in the low-fluence
64 neutron irradiation passivate the vacancy-decorated threading dislocations thus to reduce
65 the reverse leakage current of AlGaIn/GaN heterostructures^{17,18}.

66 The atomic geometries of point defects in GaN have been identified by combining the
67 experimental measurements^{13,14,19,20} and the theoretical calculations with respect to dif-
68 ferent charge states^{15,21-24}. The dominant point defect is 3+ charged N vacancy in *p*-type
69 GaN^{15,21-24}, but it might change to 3- charged Ga vacancy in *n*-type GaN^{15,22,23}. The atomic
70 structures of N and Ga interstitials have been also resolved^{19-22,25-27}. The N⁺-N(01 $\bar{1}$ 0) con-
71 figuration is recognized as the most stable N interstitial with the defect level at 1.0 eV below

72 the conduction band edge^{19,20}. Ga interstitials are experimentally detected to possess C_{3v}
73 symmetry²⁷ locating at either octahedral or tetrahedral sites, but the latter occupation is
74 excluded afterwards since the formation energy is higher by 93 meV according to the hybrid
75 DFT calculation²⁸. In addition to the thermodynamically stable configurations, the dynam-
76 ical properties of point defects have been investigated as well. The positron annihilation
77 experiment indicates that the migration barrier of N vacancy is 2.5 eV²⁹ in line with the
78 calculated value of 2.6 eV³⁰ (2.7 eV³¹) for 3+ charged state, while Ga vacancy migrates with
79 a barrier of 1.5 eV³² in agreement with the theoretical value of 1.9 eV^{21,30} for 3- charged
80 state. As for N and Ga interstitials, the minimum migration barrier is predicted to be 1.4
81 eV³⁰ (1.7 eV³¹) for 3+ charged N interstitials, and it is 0.7 eV³¹ (0.9 eV³⁰) for 3+ charged
82 Ga octahedral interstitials in good agreement with the experimental value of 0.7 eV for Ga
83 migration in the irradiated GaN²⁷.

84 Besides point defects, threading dislocations are also present in GaN due to the lattice
85 mismatch and the different thermal expansion between epilayers and substrates. With the
86 presence of dislocations, the long-range elastic field can drive point defects to move direc-
87 tionally for the release of their local strain. This causes interstitials and vacancies to migrate
88 toward the tensile and compressive regions of dislocations, respectively. Therefore, the dis-
89 locations act as sinks for point defects to form complex defective structures analogous to
90 the dislocation-induced phase segregation in Ga(In, Al)N alloys³³⁻³⁶. In turn, point defects
91 influence the structural and electronic properties of threading dislocations. The vacancy-
92 decorated a -edge dislocations form the reverse leakage current channels in AlGaIn/GaN
93 heterostructures^{17,18}. Concerning the presence of external elastic field, the metastable or
94 excited configurations of point defects are important to identify the diffusion pathways with
95 lower migration barrier as intermediate states^{19,25}. Thereby the systematical analysis of
96 native point defects in GaN is crucial to clarify the interaction properties of point defects
97 with threading dislocations in order to elucidate the degradation mechanisms of GaN-based
98 devices.

99 The defect systems of interest contain a large amount of atoms. In order to balance
100 reliability and efficiency, (semi-)empirical interatomic potentials are extremely suitable for
101 the theoretical simulations. So far, several interatomic potentials have been developed for
102 GaN, such as Stillinger-Weber (SW) potential^{37,38}, polymorphic Stillinger-Weber potential³⁹,
103 Modified Embedded-Atom Method (MEAM) potential⁴⁰, analytical bond-order potential⁴¹

104 *etc.*. They have been applied to simulate the atomic structures of defects and the dynamical
105 processes of irradiation damage. With the parameterized Ga-Ga and N-N interactions³⁷, the
106 SW potential was used to investigate threading dislocations^{38,42,43}, partial dislocations⁴⁴, tilt
107 grain boundaries^{45,46} and point defect²⁶ *etc.*. However, the version of SW potential³⁸ used
108 in the literature²⁶ fails to reproduce the most stable configurations of N and Ga interstitials.
109 Considering the successful applications in the studies of defective structures, it is worth
110 optimizing the SW potential continually to improve its performance in the simulation of
111 point defects. As an alternative option, the MEAM potential for GaN developed by Do
112 *et al.*⁴⁰ could additionally describe the elemental states of Ga and N atoms so that it is
113 inherently applicable to simulate the growth process involving the dissolution or association
114 of N₂ molecule and the clustering of Ga atoms. However, the assessment of MEAM potential
115 is still lacking to describe point defects or threading dislocations in wurtzite GaN. On the
116 other hand, the atomic structures of native point defects in GaN have not been completely
117 searched out yet as presented in the next sections.

118 Therefore, the purpose of this work is two folds. (1) To further modify the SW potential
119 for GaN and perform the assessments of it in comparison with the MEAM counterpart. (2)
120 To systematically explore the native point defects and their complexes in GaN using both
121 potentials. Subsequently, the structural and thermodynamical properties of defective struc-
122 tures are refined by DFT calculations, which thus serves as the assessment of two potentials
123 in the simulation of systems containing point defects. Accordingly, in Sec. II, the formalism
124 of SW potential is described in brief with the presence of the corresponding parameters for
125 Ga-N, Ga-Ga and N-N interactions in triplet format. The default computational details are
126 introduced here, and the exceptions will be stated whenever they are implemented. In Sec.
127 III, the fundamental properties of wurtzite GaN including the lattice parameters, elastic con-
128 stants and phonon dispersion are reproduced by the SW and MEAM potentials followed by
129 their assessments of inversion domain boundaries and multiple crystallographic structures.
130 Sec. IV presents the systematical exploration of the native point defects and their complexes
131 using the two potentials with the benchmark of DFT calculations, the atomic structures and
132 the thermal stability of which are discussed in details. Finally, the summaries are given in
133 Sec. V.

134 **II. POTENTIAL MODEL AND COMPUTATIONAL DETAILS**

135 **A. Formalism of Stillinger-Weber potential**

In the Stillinger-Weber potential⁴⁷ the total energy E of N -atom system is truncated to the two-body ϕ_2 and three-body ϕ_3 terms restricted to the second nearest neighbors:

$$E(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) = \sum_{\substack{i,j \\ (i < j)}}^N \phi_2(\mathbf{r}_i, \mathbf{r}_j) + \sum_{\substack{i,j,k \\ (i \neq j; j < k)}}^N \phi_3(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k),$$

136 where

$$137 \quad \phi_2(\mathbf{r}_i, \mathbf{r}_j) = A\epsilon \left[B \left(\frac{r_{ij}}{\sigma} \right)^{-p} - \left(\frac{r_{ij}}{\sigma} \right)^{-q} \right] \exp \left(\frac{1}{r_{ij}/\sigma - a} \right), \quad (1)$$

$$138 \quad \phi_3(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) = \lambda\epsilon (\cos \theta_{jik} + 1/3)^2 \exp \left(\frac{\gamma}{r_{ij}/\sigma - a} \right) \exp \left(\frac{\gamma}{r_{ik}/\sigma - a} \right). \quad (2)$$

139 Here r_{ij} is the distance between i, j atoms. ϵ and σ are the energy and distance scales, respectively. a represents the cut-off distance in which the interatomic interactions are counted. θ_{jik} is the angle between \mathbf{r}_{ji} and \mathbf{r}_{ik} bond vectors subtended at the vertex i atom. A, B, λ and γ are the corresponding bond strength factors.

143 Besides the regular terms of N-Ga-Ga (Ga-N-N), N-N-N and Ga-Ga-Ga, the N-Ga-N
144 (N-N-Ga) and Ga-Ga-N (Ga-N-Ga) triplets are *explicitly* considered and optimized in the
145 current version of SW potential. In the beginning, the parameters of the four regular triplets
146 take the values presented by Béré *et al.*³⁸, the geometric mean of which is assigned to the
147 other four triplets as the initial values. The parameters $\gamma = 1.2$, $p = 4$ and $q = 0$ are
148 preserved as usual^{37,38}. Then the other potential parameters are further modified⁴⁸ so that
149 the lattice parameters, elastic constants of wurtzite GaN as well as the atomic geometries
150 and the formation energy of two inversion domain boundaries (Holt⁴⁹ and Star types⁵⁰)
151 are reproduced as well as possible. The optimized results will be presented with other
152 assessments in Sec. III. In the optimization procedure, the energy scale ϵ is reassigned by
153 the one-fourth of cohesive energy (-9.058 eV) of GaN⁵¹ for N-Ga bonds, the bond energy
154 (1.731 eV) of N-N single bond⁵² for N-N bonds, and the half of cohesive energy (-2.810 eV)
155 of Ga metals⁵³ for Ga-Ga bonds, respectively. Finally, the whole set of potential parameters
156 are presented in Table I for GaN.

157 B. Computational details

158 In this paper, all Molecular Statics (MS) and Molecular Dynamics (MD) simulations
159 based on the SW and MEAM potentials are carried out by Large-scale Atomic/Molecular
160 Massively Parallel Simulator (LAMMPS) package⁵⁴. By default, the energy and force toler-
161 ance is respectively set to 10^{-15} eV and 10^{-12} eV/Å for the structural relaxation in MS sim-
162 ulation. The Density Functional Theory (DFT) calculations are implemented using Vienna
163 Ab initio Simulation Package (VASP) code⁵⁵. The electron-ionic interaction is described by
164 the Projector Augmented Wave (PAW) method⁵⁶, where $2s^2$, $2p^3$ electrons of nitrogen and
165 $3d^{10}$, $4s^2$ and $4p^1$ electrons of gallium atoms are taken as the valence states, respectively.
166 The Perdew-Burke-Ernzerhof (PBE) scheme⁵⁷ is used for the exchange and correlation func-
167 tional. The energy tolerance is 10^{-6} eV in the electronic iteration and the force is minimized
168 to 0.01 eV/Å during the geometry optimization. The conjugate gradient algorithm and the
169 periodic boundary condition are applied in the calculations of empirical potential and DFT
170 method. The geometries of supercells and the atomic positions are relaxed fully.

171 III. ASSESSMENT OF EMPIRICAL POTENTIALS

172 A. Lattice parameters

173 The primitive cell of wurtzite GaN is illustrated in Fig. 1. As seen in Table II, both the
174 SW and MEAM potentials reasonably reproduce the lattice constants a and c of wurtzite
175 GaN. But the ratio of c/a and the internal structural parameter u deviate from the exper-
176 imental values⁵⁸ due to the lack of long range interactions over the third-nearest neighbors
177 in the two empirical potentials, such as electrostatic effect.

178 B. Elastic properties

179 The elastic parameters are calculated by the SW and MEAM potentials in comparison
180 with the Antenna-Transmission Acoustic-Resonance (ATAR) experimental data⁵⁹ as pre-
181 sented in Table II. The elastic constants are directly calculated according to their definition
182 $C_{ij} = \partial \mathbf{F}_i / \partial \mathbf{x}_j$, where \mathbf{F}_i is the force along \mathbf{x}_i axis on the \mathbf{x}_j plane. For the wurtzite
183 phase of GaN, there are five independent elastic constants⁹ as $C_{11} = C_{22}$, C_{33} , $C_{12} = C_{21}$,

184 $C_{13} = C_{31} = C_{23} = C_{32}$, $C_{44} = C_{55}$, and $C_{66} = (C_{11} - C_{12})/2$. The bulk modulus is cal-
 185 culated as $B = \frac{C_{33}(C_{11}+C_{12})-2(C_{13})^2}{C_{11}+C_{12}+2C_{33}-4C_{13}}$. The maximum deviation from the experimental data is
 186 8.8% (C_{13}) for the SW potential but 14.0% (C_{12}) high for the MEAM counterpart. The elas-
 187 tic constants and bulk modulus calculated by the SW potential are also in good agreement
 188 with DFT calculations⁴⁰. Therefore, the SW potential presented in Table I can properly
 189 describe the mechanical properties of GaN.

190 C. Phonon dispersion

191 The phonon dispersion characterizes the collective vibration of atoms around their equi-
 192 librium lattice positions with respect to the wave vectors. The branches of phonon dispersion
 193 reflect the specific features of crystal structure and the interatomic interaction⁶⁰. In order
 194 to investigate the thermal properties of GaN and the dynamical properties of lattice de-
 195 fects correspondingly, it is necessary to assess the performance of empirical potentials to
 196 reproduce the phonon dispersion of crystals.

197 The phonon dispersion of GaN is calculated using the FixPhonon module^{61,62} in LAMMPS.
 198 In this method, the elements of dynamical matrix are constructed directly from the long-
 199 time trajectories of atoms in the MD simulations, since the ensemble average of the second
 200 moment of atomic displacements at a finite temperature is equal to the lattice Green's
 201 function coefficient, which is inversely proportional to the force constant according to the
 202 fluctuation-dissipation theorem⁶³.

203 Since GaN is stiff with the high elastic constants as shown in Table II, a large supercell
 204 is critically required for the computational convergence of dynamical matrices, the size of
 205 which is optimized to be $14a \times 14a \times 12c$ ⁶⁴ along the $[2\bar{1}\bar{1}0]$, $[\bar{1}2\bar{1}0]$ and $[0001]$ directions,
 206 respectively. Totally 9408 atoms are contained. The MD simulations are implemented in the
 207 microcanonical ensemble with the Langevin thermostat⁶⁵, in which the damp parameter is
 208 0.2, and the time step is set to 2 fs. The system pressure is adjusted to 1 atmosphere with the
 209 temperature of 0.1 K. The atomic displacements are recorded for 20 ns after the time of 1 ns
 210 for the thermal equilibration of system. The phonon dispersion and the Phonon Density Of
 211 States (PDOS) are subsequently calculated using the auxiliary post-processing code *phana*⁶²
 212 and shown in Fig. 2 for the SW and MEAM potentials. The phonon frequencies at some
 213 high-symmetry k points in Brillouin zone are listed in Table III in comparison with the

214 experimental data of Inelastic X-Ray Scattering (IXS) at 300 K⁶⁶ and of Raman scattering
215 at 6K⁶⁷. The results of density-functional perturbation theory (DFPT) calculations⁶⁶ are
216 also listed for comparison.

217 As shown in Fig. 2, both SW and MEAM potentials yield twelve phonon dispersion
218 branches for the 4-atom primitive cell of wurtzite GaN (Fig. 1). In the acoustic regime,
219 the curve profile, energy spanning and symmetry degeneracies of the longitudinal-acoustic
220 (LA) and transverse-acoustic (TA) branches are consistent with DFPT calculation (Fig. 2
221 in Reference⁶⁶) very well, but agree fairly for the profile of longitudinal-optical (LO) and
222 transverse-optical (TO) branches in the high energy regime. Specially along the Γ -A direc-
223 tion, the energy interval between LO and TO phonon calculated by two empirical potentials
224 is much smaller than that of DFPT calculation. Typically, at A point the frequency interval
225 is estimated to be 5.3 THz in DFPT calculation⁶⁶, while it is only about 0.6 THz for the
226 SW and MEAM potentials. The deviation of the optical phonon branches could originate
227 from the lack of long range electrostatic interaction in the used empirical potentials, which
228 is characteristic in polar materials and results in the LO-TO splitting⁶⁸. In details, when
229 the wave vector is along the Γ -A direction, the spontaneous polarization contributes to the
230 interatomic interaction parallel to the \mathbf{c} axis more than the components within the \mathbf{c} plane.
231 In this regard, the extra polarization effect causes an additional force for the longitudinal
232 mode in comparison to the transverse mode for the atomic vibration within Ga-N pairs,
233 which yields higher frequency for LO phonon than TO phonon. In addition, the phonon
234 frequencies calculated by the SW and MEAM potentials are broadly higher than either the
235 experimental data or DFPT calculations throughout the entire Brillouin zone as indicated
236 in Table III. Taking the results of DFPT calculation⁶⁶ as reference, the maximum deviation
237 happens along three TO modes (A1(TO), E1(TO) and E2) at Γ point, which is up to 7.5
238 THz (31 meV) and 8.5 THz (35 meV) for the SW and MEAM potentials, respectively. The
239 values are rather smaller than the energy scale interested in the following investigation of
240 point defects, and the SW and MEAM potentials are thus reasonably acceptable to describe
241 interatomic interactions in GaN.

242 **D. Inversion domain boundaries**

243 Besides the regular Ga-N bonds, the interactions of Ga-Ga and N-N pairs are considered
 244 in the SW and MEAM potentials as well. A typical planar defect of Inversion Domain
 245 Boundary (IDB) is selected for the optimization and assessment of the corresponding poten-
 246 tial parameters. The IDB structures have two atomic configurations. One is the Holt-type
 247 denoted as IDB^H⁴⁹, where Ga and N atoms are interchanged to occupy the opponent sites
 248 in their sublattice to form a boundary along the $\{01\bar{1}0\}$ plane as shown in Fig. 3 (Left).
 249 The generated Ga-Ga and N-N bonds are alternately arranged along the boundary of IDB^H.
 250 The other configuration of IDB is denoted as IDB*^{50,69,70}, which is formed by translating one
 251 side of IDB^H by $c/2$ along the $[0001]$ direction to avoid Ga-Ga and N-N bonds. Then 4-fold
 252 and 8-fold atomic rings are formed in pair along the boundary as shown in Fig. 3 (Right).

253 In order to preserve the periodicity, two same IDB boundaries are introduced into one
 254 supercell, which are arranged reversely with the same interval along the $[01\bar{1}0]$ direction. The
 255 size of supercell is taken as $8\sqrt{3}a \times 1c \times 2a$ along the $[01\bar{1}0]$, $[0001]$ and $[2\bar{1}\bar{1}0]$ directions,
 256 respectively. 128 atoms are contained inside. The formation energy E_{form} of IDB structures
 257 is defined as $E_{\text{form}} = (E_{\text{IDB}} - E_0)/2S$, where E_{IDB} and E_0 are the total energy of the defective
 258 and perfect structures, respectively. S is the area of the single interface. The relaxed bond
 259 length of N-N and Ga-Ga bonds in IDB^H and the averaged bond length of N-Ga bonds in
 260 4-fold atomic rings of IDB* are presented in Table IV in comparison with DFT calculations⁵⁰.

261 The formation energies of IDB* and IDB^H calculated by the SW and MEAM poten-
 262 tials indicate that IDB^H is less stable than IDB* configuration in agreement with DFT
 263 calculations⁵⁰. The value of formation energy of IDB* calculated by the MEAM potential
 264 is closer to the result of DFT calculation than the SW potential. But the atomic geome-
 265 tries of two IDB structures relaxed by the SW potential are more consistent with DFT
 266 calculations as indicated by the more similar N-N and Ga-Ga bond length in IDB^H and
 267 by the identically rectangular shape of 4-fold atomic ring in IDB* structure (Fig. 1 (b) in
 268 Reference⁵⁰), respectively. The MEAM potential generates shorter N-N and Ga-Ga bonds
 269 in IDB^H and a tilted shape of 4-fold atomic ring in IDB* structures. The shorter N-N and
 270 Ga-Ga bonds in IDB^H could be attributed to the shorter equilibrium distance (1.10 Å) and
 271 higher cohesive energy (4.88 eV) of N-N dimer in the MEAM potential⁷¹. In the relaxation
 272 of IDB^H structure, the N-N and Ga-Ga bonds competitively deform to minimize the system

273 energy, which expands N-N but compresses Ga-Ga bonds, and the distortion is dominantly
 274 suffered by Ga-Ga bonds. However, the equilibrium distance of N-N and Ga-Ga bonds is
 275 respectively 1.47 Å and 2.39 Å in the SW potential, close to the N-Ga bond length of 1.95 Å.
 276 Accordingly, the interface of IDB^H structure is distorted fairly when using the SW potential.
 277 The tilted 4-fold atomic rings in IDB* structure could be also ascribed to the parameters of
 278 MEAM potential, in which the minimum screening cutoff C_{min} is 2.0 for N-N pairs and 1.4
 279 for Ga-Ga pairs⁴⁰. Beginning with the rectangular 4-fold atomic rings, more atoms (either
 280 Ga or N atoms) will be involved into the screening effect when the diagonal N-N interaction
 281 is accounted than the case of Ga-Ga interaction, so that the N-N bonds are weakened much
 282 more than the diagonal Ga-Ga bonds to cause the shear deformation during the structural
 283 relaxation.

284 Therefore, the SW and MEAM potentials are applicable to the defective structures con-
 285 taining N-N and Ga-Ga bonds besides the regular N-Ga ones. The former would be good at
 286 the structural description while the latter would be more precise to the energy calculation.

287 E. Multiple crystallographic structures

288 In order to evaluate the transferability of the SW and MEAM potentials with respect
 289 to different atomic environments, the wurtzite, zinc-blende and rocksalt structures with
 290 different coordinate numbers are considered. In the wurtzite and zinc-blende phases, each
 291 cation (anion) has 4 neighboring anions (cations) in a tetrahedral coordination, whereas
 292 in the rocksalt phase, each cation (anion) binds 6 neighboring anions (cations) to form an
 293 octahedral structure. The binding energies as function of volume per formula unit (N-Ga)
 294 in the three lattices are plotted in Fig. 4. The wurtzite and zinc-blende phases can not be
 295 distinguished by either SW or MEAM potentials since the cutoff radius is restricted to the
 296 second nearest neighbors in GaN. Nevertheless, the wurtzite and zinc-blende phases are more
 297 stable than rocksalt in agreement with the experiment⁷² and DFT calculation⁷³. Moreover,
 298 the rocksalt phase is stable under high pressure. The equilibrium transition pressure P_T for
 299 the wurtzite-rocksalt phase transition is 52.2 GPa in experiment⁷² and 44.5 GPa as predicted
 300 by DFT calculation⁷³. P_T could be estimated by the common tangent construction of two
 301 energy curves in Fig. 4. Obviously, *only* the MEAM potential could qualitatively describe
 302 the wurtzite-rocksalt phase transition even the calculated value of P_T is fairly high (106

303 GPa). The SW potential fails to predict the phase transition since the three-body term
 304 is rigid to fix the tetrahedral configuration determined by sp^3 hybridization. Indeed, any
 305 distortion from the ideal structure increases the system energy, so the energy curves of
 306 wurtzite and rocksalt phases can not cross within a reasonable strain range and no sound
 307 value could be estimated for P_T .

308 Therefore, the MEAM potential is more transferable to treat the structural change in
 309 regard to the variation of coordinate numbers, but the SW potential is still applicable to
 310 describe the atomic structures not far away from the tetrahedral type.

311 IV. EXPLORATION OF NATIVE POINT DEFECTS AND THEIR 312 COMPLEXES

313 A. Native point defects

314 With the SW potential parameterized in Table I, we explore the atomic structures of
 315 point defects in wurtzite GaN in comparison with the calculations based on the MEAM
 316 potential. The configurations of native point defects include three types, namely vacancy,
 317 antisite and interstitial. We first investigate the single point defects and then the point
 318 defect complexes made up of two point defects.

319 The native point defects and their complexes are modeled using a supercell with the size
 320 of $5a \times 3\sqrt{3}a \times 3c$ along the $[2\bar{1}\bar{1}0]$, $[01\bar{1}0]$ and $[0001]$ direction, respectively. 360 atoms are
 321 contained for the perfect structure. As illustrated in Fig. 5, the possible atomic structures of
 322 native point defects are constructed as follows, and the split interstitials are denoted accord-
 323 ing to the orientation of dumbbells: (1) Anion (Cation) vacancies, V_N (V_{Ga}), where a N (Ga)
 324 atom is removed from GaN bulk. (2) Anion (Cation) antisites, N_{Ga} (GaN), where the host Ga
 325 (N) atom is replaced by a N (Ga) atom. (3) Anion (Cation) octahedral/tetrahedral intersti-
 326 tials, N_{Oct} (Ga_{Oct})/ N_{Tet} (Ga_{Tet}), where a N (Ga) atom occupies the octahedral/tetrahedral
 327 site in the shuffle plane as labeled in Fig. 1 (b). (4) $\langle 2\bar{1}\bar{1}0 \rangle$ -oriented split interstitials,
 328 $N^+-N\langle 2\bar{1}\bar{1}0 \rangle$ ($Ga^+-N\langle 2\bar{1}\bar{1}0 \rangle$) and $N^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$ ($Ga^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$), where one N (Ga) intersti-
 329 tial (labeled by the symbol of '+' on the right superscript) and a host N or Ga atom share
 330 the same lattice position to form a dumbbell with the axis along the $\langle 2\bar{1}\bar{1}0 \rangle$ direction in
 331 the glide plane as labeled in Fig. 1 (b). Similarly, (5) $\langle 01\bar{1}0 \rangle$ -oriented split interstitials,

332 $N^+-N\langle 01\bar{1}0 \rangle$ ($Ga^+-N\langle 01\bar{1}0 \rangle$) and $N^+-Ga\langle 01\bar{1}0 \rangle$ ($Ga^+-Ga\langle 01\bar{1}0 \rangle$), where one N (Ga) intersti-
 333 tial and a host N or Ga atom share the same lattice position to form a dumbbell with the
 334 axis along the $\langle 01\bar{1}0 \rangle$ direction in the glide plane. Due to the lack of mirror symmetry about
 335 the $\{01\bar{1}0\}$ plane, the polarized N-Ga dumbbells are not equivalent when they orientate
 336 along the $\langle 01\bar{1}0 \rangle$ and its *opposite* directions, so the other two configurations are constructed
 337 by reversing the direction of N-Ga dumbbells, which are denoted as $Ga^+-N\langle 01\bar{1}0 \rangle$ -o and
 338 $N^+-Ga\langle 01\bar{1}0 \rangle$ -o, respectively. Totally eighteen different candidates are built as the initial
 339 atomic configurations. The defective structures are relaxed by the SW and MEAM poten-
 340 tials. Some of initial configurations convert into the identical atomic structures after the
 341 structural relaxation. The obtained atomic structures are further refined by the relaxation
 342 of DFT calculations. The computational details have been presented in Sec. II B.

343 The atomic structures relaxed by the SW, MEAM potentials and DFT calculations are
 344 shown in Figs. 6, 7 and 8, respectively. The atomic configurations of V_N , V_{Ga} and Ga_N are
 345 preserved after the structural relaxation of three methods despite the observable deformation
 346 of Ga-Ga bonds in Ga_N , which are expanded by +15.6%, +11.8% and +17.9% in average
 347 over the ideal N-Ga bonds in the relaxation of SW, MEAM potentials and DFT calculation,
 348 respectively. As for N_{Ga} antisite, the atomic configuration is preserved in the relaxation
 349 of SW potential (Fig. 6 (c)), while the N atom shifts upward to occupy the tetrahedral
 350 site when the MEAM potential is applied as shown in Fig. 7 (c). Close to the result of
 351 SW potential, Fig. 8 (c) indicates that the N atom of the DFT-relaxed N_{Ga} antisite shifts
 352 aside slightly to form a $N^+-N\langle 01\bar{1}0 \rangle$ dumbbell in the glide space instead of the reported
 353 $N^+-N\langle 0001 \rangle$ split interstitial²⁶. The N-N bond length is 1.24 Å in the formed N-N dumbbell.

354 The initial configurations of N interstitials illustrated in Fig. 5 are not all preserved
 355 in the structural relaxations of three methods. Based on the calculations of SW po-
 356 tential, $N^+-N\langle 01\bar{1}0 \rangle$ -g, $N^+-N\langle 2\bar{1}\bar{1}0 \rangle$ and $N^+-N\langle 01\bar{1}0 \rangle$ -e split interstitials are obtained as
 357 shown in Figs. 6 (e), (f) and (g), respectively. $N^+-N\langle 01\bar{1}0 \rangle$ -g and $N^+-N\langle 01\bar{1}0 \rangle$ -e denote
 358 the ground and excited states of $N^+-N\langle 01\bar{1}0 \rangle$ split interstitials as proposed in Reference¹⁹.
 359 $N^+-N\langle 01\bar{1}0 \rangle$ -g split interstitial is relaxed from N_{Tet} and $N^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$ initial configurations,
 360 while $N^+-N\langle 01\bar{1}0 \rangle$ -e is relaxed from $N^+-N\langle 01\bar{1}0 \rangle$ and $N^+-Ga\langle 01\bar{1}0 \rangle$ -o ones. In $N^+-N\langle 01\bar{1}0 \rangle$ -g
 361 split interstitial, the N-N dumbbell is almost parallel to the $\{03\bar{3}4\}$ plane (Refer to the
 362 inset of Fig. 8.), whereas the N-N dumbbell turns to be nearly perpendicular to the $\langle 03\bar{3}4 \rangle$
 363 plane and forms a small tetrahedron with two neighboring Ga atoms in $N^+-N\langle 01\bar{1}0 \rangle$ -e split

364 interstitial. The bond length of N-N dumbbell is of 1.35 Å in $N^+-N\langle 01\bar{1}0\rangle$ -g, 1.45 Å in
 365 $N^+-N\langle 2\bar{1}\bar{1}0\rangle$ and 1.35 Å in $N^+-N\langle 01\bar{1}0\rangle$ -e split interstitials, respectively, all of which consid-
 366 erably agree with the values relaxed in the following DFT calculations. As shown in Fig. 6
 367 (h), the configuration of N_{Oct} interstitial is preserved in the relaxation of SW potential, and
 368 $N^+-Ga\langle 01\bar{1}0\rangle$ split interstitial is unstable and convert into N_{Oct} as well, which is different
 369 from the relaxations of MEAM potential and DFT calculations.

370 Besides four N interstitials ($N^+-N\langle 01\bar{1}0\rangle$ -g, $N^+-N\langle 2\bar{1}\bar{1}0\rangle$, $N^+-N\langle 01\bar{1}0\rangle$ -e and N_{Oct}) in com-
 371 mon with the results of SW potential, the MEAM potential additionally predicts N tetra-
 372 hedral interstitial N_{Tet} and $N^+-Ga\langle 01\bar{1}0\rangle$ split interstitial, all of which are shown in Figs. 7
 373 (e)-(j). After the structural relaxations of MEAM potential, $N^+-N\langle 01\bar{1}0\rangle$ and $N^+-Ga\langle 2\bar{1}\bar{1}0\rangle$
 374 initial configurations convert into $N^+-N\langle 01\bar{1}0\rangle$ -g split interstitial, while $N^+-N\langle 01\bar{1}0\rangle$ -e split
 375 interstitial is relaxed from $N^+-Ga\langle 01\bar{1}0\rangle$ -o initial configuration. It should be stressed that in
 376 $N^+-N\langle 01\bar{1}0\rangle$ -g split interstitial relaxed by the MEAM potential, the N-N dumbbell is instead
 377 almost parallel to the $\{0\bar{3}38\}$ plane (Refer to the inset of Fig. 8.) as seen in Fig. 7 (e),
 378 which is different from the results of SW potential and the latter DFT calculation. In N_{Tet}
 379 interstitial, the N atom occupied the tetrahedral site binds with the host N and Ga atoms
 380 to form a straight Ga-N-N-Ga atomic chain along the $[0001]$ direction as shown in Fig. 7 (j).
 381 The bond length of N-N dumbbell is of 1.11 Å in $N^+-N\langle 01\bar{1}0\rangle$ -g, 2.29 Å in $N^+-N\langle 2\bar{1}\bar{1}0\rangle$, 1.13
 382 Å in $N^+-N\langle 01\bar{1}0\rangle$ -e split interstitials and 1.10 Å in N_{Tet} , respectively. The N-Ga dumbbell
 383 in $N^+-Ga\langle 01\bar{1}0\rangle$ split interstitial is in tensile state with the bond length of 1.96 Å longer
 384 than the ideal value of 1.94 Å for N-Ga bonds in the perfect structure.

385 After the refinement of DFT calculations, $N^+-N\langle 01\bar{1}0\rangle$ -g, $N^+-N\langle 2\bar{1}\bar{1}0\rangle$, $N^+-N\langle 01\bar{1}0\rangle$ -e and
 386 $N^+-Ga\langle 01\bar{1}0\rangle$ four N split interstitials are obtained as shown in Figs. 8 (e)-(h). N_{Oct} and N_{Tet}
 387 interstitials are unstable and convert into $N^+-N\langle 01\bar{1}0\rangle$ -g split interstitial. Nevertheless, the
 388 structural stability of N_{Oct} is still in doubt even on the same level of the first-principles calcu-
 389 lations. For instance, it has been produced when the hybrid quantum mechanical/molecular
 390 mechanical (QM/MM) approach is applied with either B97-2 or BB1k exchange-correlation
 391 functionals²³. The corresponding discussion with respect to the different computational
 392 schemes is beyond the scope of this work thus to be omitted here. Similar to the result
 393 of SW potential (Fig. 6 (e)), the N-N dumbbell in the DFT-relaxed $N^+-N\langle 01\bar{1}0\rangle$ -g split
 394 interstitial is nearly parallel to the $\{0\bar{3}\bar{3}4\}$ plane as shown in Fig. 8 (e), and each N atom
 395 of N-N dumbbell binds two neighboring Ga atoms. The bond length of N-N dumbbell is

396 of 1.35 Å in agreement with the reported value²⁵. In the DFT-relaxed N⁺-N⟨01 $\bar{1}$ 0⟩-e split
 397 interstitial (Fig. 8 (g)), the N-N dumbbell is almost perpendicular to the {03 $\bar{3}$ 4} plane
 398 to form a small tetrahedron with two neighboring Ga atoms as well, the bond length of
 399 which is 1.37 Å. Besides two N⁺-N⟨01 $\bar{1}$ 0⟩ split interstitials, N⁺-N⟨2 $\bar{1}$ $\bar{1}$ 0⟩ and N⁺-Ga⟨01 $\bar{1}$ 0⟩
 400 configurations are newly explored in this study. In N⁺-N⟨2 $\bar{1}$ $\bar{1}$ 0⟩ split interstitial, the N-N
 401 dumbbell locates in the glide space and exactly arrows along the ⟨2 $\bar{1}$ $\bar{1}$ 0⟩ direction with the
 402 bond length of 1.40 Å. It should be pointed out that Gao *et al.*²⁶ have improperly denoted
 403 N⁺-N⟨01 $\bar{1}$ 0⟩-g as N⁺-N⟨2 $\bar{1}$ $\bar{1}$ 0⟩ split interstitial, since the description of this split interstitial
 404 in their literature²⁶, that each N atom of N-N dumbbell forms two bonds with the surround-
 405 ing Ga atoms, is consistent with N⁺-N⟨01 $\bar{1}$ 0⟩-g rather than N⁺-N⟨2 $\bar{1}$ $\bar{1}$ 0⟩ split interstitial.
 406 In N⁺-Ga⟨01 $\bar{1}$ 0⟩ split interstitial, the N-Ga dumbbell tilts by about 20° from the {0001}
 407 towards the {03 $\bar{3}$ 4} plane as shown in Fig. 8 (h). The N-Ga dumbbell is in compressive
 408 state with the bond length of 1.80 Å shorter than the ideal N-Ga bonds in the perfect
 409 structure (1.96 Å along the *c* axis and 1.95 Å for the other three bonds in the *c* plane).
 410 Therefore, the structural description of N-N bonds in N split interstitials by the SW poten-
 411 tial is more consistent with DFT calculations than the MEAM counterpart, which is similar
 412 to the investigation of IDB^H structure in Sec. III D.

413 In the exploration of Ga interstitials, Ga_{Oct}, Ga_{Tet} interstitials and Ga⁺-Ga⟨01 $\bar{1}$ 0⟩-e,
 414 Ga⁺-Ga⟨2 $\bar{1}$ $\bar{1}$ 0⟩ split interstitials are produced by the SW potential as shown in Figs. 6 (i)-(l).
 415 Ga⁺-Ga⟨01 $\bar{1}$ 0⟩-e split interstitial is relaxed from the initial configurations of Ga⁺-N⟨01 $\bar{1}$ 0⟩
 416 and Ga⁺-Ga⟨01 $\bar{1}$ 0⟩. Since the Ga-Ga dumbbell is nearly perpendicular to the {03 $\bar{3}$ 4} plane
 417 and binds with two neighboring N atoms to form a small tetrahedron analogous to the
 418 structure of N⁺-N⟨01 $\bar{1}$ 0⟩-e split interstitial, the index ‘-e’ is added in the denotation of
 419 Ga⁺-Ga⟨01 $\bar{1}$ 0⟩-e split interstitial. The bond length of Ga-Ga dumbbell is 2.26 Å. The
 420 Ga⁺-N⟨01 $\bar{1}$ 0⟩-o and Ga⁺-N⟨2 $\bar{1}$ $\bar{1}$ 0⟩ initial configurations are unstable and convert into Ga_{Oct}
 421 after the structural relaxation. Different from the case of N_{Tet}, the configuration of Ga_{Tet} is
 422 stable, where the Ga interstitial atom pushes the underlying host Ga-N pair downwards to
 423 bind with the below Ga atom, then a straight N-Ga-Ga-N-Ga atomic chain is formed along
 424 the *c* axis, which has been denoted as the bridge-bond geometry in the literature⁷⁴. The
 425 interatomic distance of the N-Ga-Ga-N-Ga atomic chain is of 2.11 Å for the Ga-Ga pair
 426 and 1.80 Å for the host N-Ga pair. In Ga⁺-Ga⟨2 $\bar{1}$ $\bar{1}$ 0⟩ split interstitial, the Ga-Ga dumbbell
 427 binds with two neighboring host Ga atoms in the same glide plane to form a curved Ga-Ga-

428 Ga-Ga atomic chain²⁶, and the mirror symmetry is preserved about the $\{2\bar{1}\bar{1}0\}$ plane same
 429 as $N^+-N\langle 2\bar{1}\bar{1}0 \rangle$ split interstitial. The interatomic distance of the metal chain is of 2.40 Å,
 430 2.31 Å and 2.40 Å, respectively.

431 Similar to the case of N interstitials, the MEAM potential predicts more configurations of
 432 Ga interstitials than the SW counterpart, which include Ga_{Oct} interstitial, $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -e,
 433 $Ga^+-N\langle 01\bar{1}0 \rangle$ $Ga^+-N\langle 01\bar{1}0 \rangle$ -o, $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -g and $Ga^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$ split interstitials as
 434 shown in Figs. 7 (k)-(p). The initial configurations of Ga_{Oct} interstitial, $Ga^+-N\langle 01\bar{1}0 \rangle$,
 435 $Ga^+-N\langle 01\bar{1}0 \rangle$ -o and $Ga^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$ split interstitials are preserved, while Ga_{Tet} interstitial is
 436 unstable and convert into $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -e split interstitial (Fig. 7 (l)). The $Ga^+-Ga\langle 01\bar{1}0 \rangle$
 437 initial configuration is relaxed into $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -g split interstitial as shown in Fig. 7
 438 (o), where the Ga-Ga dumbbell tilts to be nearly parallel to the $\{03\bar{3}4\}$ plane analogous
 439 to the structure of $N^+-N\langle 01\bar{1}0 \rangle$ -g split interstitial, so that the index ‘-g’ is added in this
 440 denotation. It should be noticed that one Ga atom of this Ga-Ga dumbbell binds with three
 441 but the other Ga atom binds with two neighboring N atoms. The bond length of Ga-Ga
 442 dumbbell is of 2.06 Å in $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -e and 1.99 Å in $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -g split interstitials,
 443 respectively. Besides the preserved candidate, the $Ga^+-N\langle 2\bar{1}\bar{1}0 \rangle$ initial configuration con-
 444 verts into $Ga^+-N\langle 01\bar{1}0 \rangle$ split interstitial as well after the structural relaxation. In the paired
 445 configurations of $Ga^+-N\langle 01\bar{1}0 \rangle$ and $Ga^+-N\langle 01\bar{1}0 \rangle$ -o split interstitials, the N-Ga dipole points
 446 to the $\langle 01\bar{1}0 \rangle$ and its reverse directions, respectively. In $Ga^+-N\langle 01\bar{1}0 \rangle$ split interstitial, the
 447 interstitial Ga atom binds with the frontal host Ga atom and pushes it forwards to form
 448 a N-Ga-Ga crowdion in the $\{2\bar{1}\bar{1}0\}$ plane, where the Ga-Ga pair is roughly parallel to the
 449 $\{0\bar{3}38\}$ plane. In the N-Ga-Ga crowdion, the interatomic distance is of 2.12 Å for the
 450 Ga-Ga pair and 1.91 Å for the N-Ga dipole in compression, whereas the N-Ga dumbbell
 451 in $Ga^+-N\langle 01\bar{1}0 \rangle$ -o split interstitial has a bond length of 1.95 Å. In line with the result
 452 of SW potential, the MEAM potential produces a curved Ga-Ga-Ga-Ga atomic chain in
 453 $Ga^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$ split interstitial, where the interatomic distance between Ga atoms is of 2.43
 454 Å, 2.08 Å and 2.43 Å, respectively.

455 The further structural relaxation of DFT calculations confirms five stable configurations
 456 of Ga interstitials, that are Ga_{Oct} , Ga_{Tet} interstitials, and $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -g, $Ga^+-N\langle 01\bar{1}0 \rangle$,
 457 $Ga^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$ split interstitials as shown in Figs. 8 (i)-(m). In Ga_{Oct} and Ga_{Tet} intersti-
 458 tials, C_{3v} symmetry is exactly preserved as discussed by Chow *et al.*²⁷. Along the straight
 459 N-Ga-Ga-N-Ga atomic chain in Ga_{Tet} interstitial, the Ga-Ga bond length is 2.21 Å and

460 the host Ga-N pair is compressed to 1.78 Å. In Ga⁺-Ga⟨01 $\bar{1}$ 0⟩-g split interstitial (Fig. 8
461 (k)), the Ga-Ga dumbbell keeps nearly parallel to the {03 $\bar{3}$ 4} plane as well with the bond
462 length of 2.16 Å. In Ga⁺-N⟨01 $\bar{1}$ 0⟩ split interstitial (Fig. 8 (l)), the N-Ga-Ga crowdion is pre-
463 served with the interatomic distance of 2.17 Å for the Ga-Ga pair and 1.77 Å for the N-Ga
464 dipole in comparison. Same to the results of SW and MEAM potentials, the DFT-relaxed
465 Ga⁺-Ga⟨2 $\bar{1}$ 10⟩ split interstitial is characterized by the Ga-Ga-Ga-Ga atomic chain as shown
466 in Fig. 8 (m). The interatomic distance is of 2.38 Å, 2.33 Å and 2.38 Å along this metal
467 chain, respectively.

468 Therefore, the atomic structures of native point defects in wurtzite GaN have been ex-
469 plored as completely as possible using the SW and MEAM potentials with the benchmark
470 of DFT calculations. In viewpoint of the potential energy landscape, the MEAM potential
471 predicts more local minima than either the SW counterpart or DFT calculations even though
472 the three methods produce a intersection set of local minima for the same atomic config-
473 urations of native point defects. For the commonly predicted point defects, the structural
474 features, typically the bond length of N-N and Ga-Ga dumbbells relaxed by the SW poten-
475 tial are more consistent with the results of DFT calculations than by the MEAM potential.
476 The distinct mappings of the energy landscapes predicted by the two potentials could be
477 understood intuitively based on their models of interatomic interaction as follows. In re-
478 gard to covalent systems, the bonding configuration and the saturation of bonding number
479 are explicitly determined by the three-body term in the SW potential (Eq. 2), while the
480 directional characteristics of covalent bonds and the influence of atomic environments are
481 implicitly described through the screening factor S_{ij} (Eq. 14 in Reference⁴⁰) in the MEAM
482 potential. The change of atomic structures is local in point defects. The SW potential could
483 sensitively count this local change once the neighboring atoms are displaced, whereas in
484 the calculation of MEAM potential, taking Ga⁺-Ga⟨01 $\bar{1}$ 0⟩ split interstitial as example, the
485 contribution of screening factor S_{ij} would vary little when the Ga-Ga dumbbell rotates since
486 the atomic environment around it is almost preserved, so that multiple stable configurations
487 may exist with respect to the different orientations of Ga-Ga dumbbell, and thus the extra
488 local minima appear in the energy landscape of MEAM potential.

489 The thermodynamical stability of neutral point defects are measured by the formation
490 energy Ω defined as²⁶

$$491 \quad \Omega = E_D - n_N \mu_N - n_{Ga} \mu_{Ga}, \quad (3)$$

492 where E_D is the total energy of defective structure containing n_N N atoms and n_{Ga} Ga atoms.
 493 μ_N and μ_{Ga} are the chemical potentials of Ga and N atoms, respectively. In the thermody-
 494 namic equilibrium, μ_N and μ_{Ga} are subject to the constraint condition of $\mu_{GaN} = \mu_N + \mu_{Ga}$
 495 where μ_{GaN} is the chemical potential of GaN. Since the chemical potentials of elemental Ga
 496 bulk and N_2 molecule are not defined in the SW potential, the relative formation energy
 497 $\Delta\Omega$ is introduced instead as $\Delta\Omega = E_D - E_{D,0}$ where $E_{D,0}$ is the total energy of reference
 498 structure. In this work, $N^+ - N\langle 01\bar{1}0 \rangle - g$ and Ga_{Oct} configurations are taken as the reference
 499 structures for N and Ga interstitials, respectively.

500 The relative formation energies $\Delta\Omega$ calculated by the SW and MEAM potentials along
 501 with the DFT results are presented in Table V for the N and Ga interstitials shown in
 502 Figs. 6, 7 and 8. In the results of DFT calculations, $N^+ - N\langle 01\bar{1}0 \rangle - g$ split interstitial is the
 503 most stable configuration thus to be taken as the ground state among N interstitials in line
 504 with the study of Bardeleben *et al*¹⁹. The new $N^+ - N\langle 2\bar{1}\bar{1}0 \rangle$ split interstitial is the second
 505 most stable configuration with 0.407 eV higher in energy, while the formation energy of
 506 $N^+ - N\langle 01\bar{1}0 \rangle - e$ split interstitial is higher by 0.447 eV than its ground state $N^+ - N\langle 01\bar{1}0 \rangle - g$ in
 507 agreement with the reported value of 0.35 eV¹⁹. The extra metastable configurations would
 508 imply new diffusion pathways for N interstitials in the process of migration and rearrange-
 509 ment. Different from DFT calculations, the SW potential predicts N_{Oct} interstitial to be
 510 the most stable configuration. Among the other three split interstitials, $N^+ - N\langle 01\bar{1}0 \rangle - g$ has
 511 the lower formation energy than either $N^+ - N\langle 2\bar{1}\bar{1}0 \rangle$ or $N^+ - N\langle 01\bar{1}0 \rangle - e$ configuration, which is
 512 consistent with the results of DFT calculations. Superior to the SW counterpart, the MEAM
 513 potential predicts $N^+ - N\langle 01\bar{1}0 \rangle - g$ split interstitial as the most stable configuration in line with
 514 the DFT calculations. But the relative formation energies of other N interstitials, specially
 515 $N^+ - N\langle 2\bar{1}\bar{1}0 \rangle$ and $N^+ - N\langle 01\bar{1}0 \rangle - e$ configurations are rather high in comparison with the results
 516 of DFT calculations, so that they will be hardly visited during the dynamic simulation of N
 517 interstitials. Regarding Ga interstitials, the DFT calculations indicate that Ga_{Oct} interstitial
 518 is the most stable configuration. And except $Ga^+ - Ga\langle 2\bar{1}\bar{1}0 \rangle$ split interstitial, the other Ga
 519 interstitials Ga_{Tet} , $Ga^+ - Ga\langle 01\bar{1}0 \rangle - g$ and $Ga^+ - N\langle 01\bar{1}0 \rangle$ have the relative formation energies
 520 less than 0.4 eV in comparison with Ga_{Oct} . The SW potential correctly predicts Ga_{Oct} to be
 521 the most stable Ga interstitial in agreement with the DFT calculations, so that this version
 522 of SW potential presented in Table I is superior to that one used in the previous work²⁶. On
 523 the other hand, rather different from the SW potential and DFT calculations, the MEAM

524 potential predicts $\text{Ga}^+-\text{Ga}\langle 01\bar{1}0 \rangle$ -e split interstitial to be the most stable configuration. But
 525 the formation energies of other Ga interstitials such as $\text{Ga}^+-\text{Ga}\langle 01\bar{1}0 \rangle$ -g and $\text{Ga}^+-\text{N}\langle 01\bar{1}0 \rangle$
 526 and $\text{Ga}^+-\text{Ga}\langle 2\bar{1}\bar{1}0 \rangle$ are reasonably consistent with the values of DFT calculations. To con-
 527 clude, neither SW nor MEAM potentials could individually perform very well to describe
 528 the interstitial point defects in GaN. However, they are complementary with respect to the
 529 structural and thermodynamical properties of those point defects, and it is worthy to use
 530 two potentials as a cross reference for the dynamical simulations of defective GaN.

531 B. Point defect complexes

532 In addition to the native point defects, their point defect complexes are further explored
 533 with respect to the interaction between two native point defects. Four point defect complexes
 534 are selected as follows. (1) Schottky, which is created by removing one N-Ga pair from GaN
 535 bulk. (2) Paired antisite $\text{N}_{\text{Ga}}\text{Ga}_{\text{N}}$, where the N and Ga atoms in one N-Ga pair occupy
 536 the opposite sites of each other. (3) Anion (Cation) octahedral Frenkel defect $\text{Frenkel}_{\text{Oct}}(\text{N})$
 537 ($\text{Frenkel}_{\text{Oct}}(\text{Ga})$), where one N (Ga) atom is moved from its equilibrium site to the nearest
 538 octahedral space to form a N (Ga) vacancy plus a N (Ga) octahedral interstitial. Similarly,
 539 (4) N (Ga) tetrahedral Frenkel defect $\text{Frenkel}_{\text{Tet}}(\text{N})$ ($\text{Frenkel}_{\text{Tet}}(\text{Ga})$), which could be initially
 540 constructed by moving one host N (Ga) atom to the nearest tetrahedral site. But the
 541 configurations are unstable and recover back to the perfect structure after the structural
 542 relaxation using either the SW potential or DFT calculation, so that they are not considered
 543 further here.

544 The relaxed atomic structures of point defect complexes are shown in Fig. 9 with respect
 545 to three methods. In details, Figs. 9 (a1)-(a3) manifest no structural collapse in the Schottky
 546 defect. There is just a slight expansion around Ga vacancy and a little shrinkage around N
 547 vacancy in the atomic structures relaxed by the DFT calculation and MEAM potential, and
 548 hardly any deformation in the calculation of SW potential. However, the DFT calculations
 549 reveal that both of the initial configurations of $\text{N}_{\text{Ga}}\text{Ga}_{\text{N}}$ and $\text{Frenkel}_{\text{Oct}}(\text{N})$ defects are unsta-
 550 ble, and they similarly convert into the point defect complexes made up of N vacancy and
 551 $\text{N}^+-\text{N}\langle 01\bar{1}0 \rangle$ -g split interstitial, where the distance between the two native point defects is
 552 approximately of $1.4a$ (Fig. 9 (b1)) and $2.4a$ (Fig. 9 (c1)) (a : lattice constant), respectively.
 553 The symbol of ‘ \oplus ’ is adopted as the linkage in point defect complexes. Thus the relaxed

554 structures of $N_{Ga}Ga_N$ and $Frenkel_{Oct}(N)$ defects are denoted as $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle-g)_{1.4a}$
 555 and $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle-g)_{2.4a}$, respectively. In contrast, the initial configuration of $N_{Ga}Ga_N$
 556 is preserved in the structural relaxation of SW and MEAM potentials as shown in Figs. 9
 557 (b2) and (b3), respectively. Furthermore, the DFT-relaxed structure (Fig. 9 (b1)) is taken
 558 as the initial configuration to be relaxed by the SW and MEAM potentials for $N_{Ga}Ga_N$
 559 defect. In the calculation of SW potential, the interstitial configuration of N-N dumbbell
 560 in $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle-g)_{1.4a}$ is replaced by a nearly horizontal N-Ga dumbbell as shown in
 561 Fig. 9 (b2'), which could be equivalently formed by rotating a vertical N-Ga pair by about
 562 90° in the perfect structure similar to the formation of Stones-Wales defect in graphite⁷⁵.
 563 Meanwhile, the initial configuration of $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle-g)_{1.4a}$ is preserved for the MEAM
 564 potential as shown in Fig. 9 (b3'). The interatomic distance of N-N pair is of 1.42 Å, 1.58
 565 Å and 1.12 Å in the atomic structures relaxed by the DFT calculation, SW and MEAM
 566 potentials, respectively. The initial configuration of $Frenkel_{Oct}(N)$ is preserved in the struc-
 567 tural relaxation of SW and MEAM potentials as well, but just only the interstitial N atom
 568 stays at an off-center site in the octahedral space restricted by six neighboring Ga atoms as
 569 shown in Figs. 9 (c2) and (c3), respectively. Similar to the case of $N_{Ga}Ga_N$, taking the DFT-
 570 relaxed structure (Fig. 9 (c1)) as the initial configuration, $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle-g)_{2.4a}$ point
 571 defect complexes are preserved in the structural relaxation of SW and MEAM potentials as
 572 shown in Figs. 9 (c2') and (c3'), respectively. The geometric features of N-N dumbbells are
 573 restored as those in the isolated $N^+-N\langle 01\bar{1}0 \rangle-g$ split interstitials shown in Fig. 8 (e) for DFT
 574 calculation, in Fig. 6 (e) for SW and in Fig. 7 (e) for MEAM potentials, respectively. The
 575 bond length of N-N dumbbell is correspondingly of 1.44 Å, 1.35 Å and 1.11 Å in the relaxed
 576 structures of DFT calculation, SW and MEAM potentials. Different from $Frenkel_{Oct}(N)$,
 577 the initial configuration of $Frenkel_{Oct}(Ga)$ is preserved for three methods as shown in Figs.
 578 9 (d1)-(d3). The interstitial Ga atom occupies the body center of octahedral space closely
 579 adjacent the Ga vacancy, and the distance between two point defects is approximately of
 580 $2\sqrt{3}/3a$ (a : lattice constant).

581 Since $n_N = n_{Ga}$ in the investigated point defect complexes, Eq. 3 could be simplified as

$$582 \quad \Omega = E_D - E_0, \tag{4}$$

583 where E_0 is the total energy of perfect structure with the same atomic number to the
 584 defective system for each elemental species. The corresponding formation energies of the

585 point defect complexes shown in Fig. 9 are listed in Table VI, which are arranged ac-
586 cording to the values of DFT calculations in ascending order. If only considering the
587 $V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g configurations instead of either $\text{Frenkel}_{\text{Oct}}(\text{N})$ or $N_{\text{Ga}}\text{Ga}_N$, the three
588 methods roughly agree for the tendency of thermodynamical stability of defective structures,
589 that Schottky is the most stable point defect complex, followed by $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g) $_{2.4a}$
590 $((V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g) $_{1.4a}$), and $\text{Frenkel}_{\text{Oct}}(\text{Ga})$ has the highest formation energy. More-
591 over, except $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g) $_{2.4a}$ the formation energies of those defects calculated by
592 the SW potential are quantitatively more consistent with the DFT results than the MEAM
593 counterpart.

594 On the other hand, the interaction between two native point defects in the point defect
595 complex is further investigated by considering the system, where two native point defects
596 locate with an infinite separation. And the corresponding formation energy could be simply
597 calculated by summing those of two isolated point defects. In order to be compatible with
598 the configurations of those point defect complexes discussed above, four paired point defects
599 are modeled as follows $V_N + V_{\text{Ga}}$, $V_N + N^+-N\langle 01\bar{1}0 \rangle$ -g, $N_{\text{Ga}} + \text{Ga}_N$ and $V_{\text{Ga}} + \text{Ga}_{\text{Oct}}$, where
600 the symbol of ‘+’ is adopted in place of ‘ \oplus ’ to signify the infinite separation between two
601 point defects. Similar to the point defect complexes, the formation energies of them are
602 calculated with Eq. 4 due to the equal atomic number of Ga and N species. The results
603 are presented in Table VI for the three methods. The DFT calculations reveal that the
604 formation energy reduces when the distance between two point defects decreases. For in-
605 stance, the formation energy reduces by 3.544 eV in the formation of Schottky defect from
606 $V_N + V_{\text{Ga}}$, and by 4.879 eV in the formation of $\text{Frenkel}_{\text{Oct}}(\text{Ga})$ from $V_{\text{Ga}} + \text{Ga}_{\text{Oct}}$. Begin-
607 ning with the paired point defect of $V_N + N^+-N\langle 01\bar{1}0 \rangle$ -g (or $N_{\text{Ga}} + \text{Ga}_N$), the formation
608 energy reduces by 0.716 eV (or 6.572 eV) when two isolated point defects move close to
609 form $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g) $_{2.4a}$. Thus the point defect complexes, at least those listed in
610 Table VI, are energetically feasible rather than dissociate into two isolated point defects.
611 The energy tendency predicted by the DFT calculations is reasonably reproduced by the
612 SW potential, that the formation energy reduces by 2.176 eV in the formation of Schottky
613 from $V_N + V_{\text{Ga}}$, by 0.016 eV or 3.363 eV in the formation of $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g) $_{2.4a}$ from
614 $V_N + N^+-N\langle 01\bar{1}0 \rangle$ -g or $N_{\text{Ga}} + \text{Ga}_N$, and by 0.025 eV in the formation of $\text{Frenkel}_{\text{Oct}}(\text{Ga})$ from
615 $V_{\text{Ga}} + \text{Ga}_{\text{Oct}}$, respectively. But the MEAM potential wrongly predicts the dissociation of
616 Schottky and $(V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g) $_{2.4a}$ $((V_N \oplus N^+-N\langle 01\bar{1}0 \rangle$ -g) $_{1.4a}$) point defect complexes

617 to be exothermic by the energy reduction of 0.034 eV for the former and of 0.024 eV (0.072
618 eV) for the latter, respectively. Therefore, the point defect complexes including Schottky,
619 $(V_N \oplus N^+ - N\langle 01\bar{1}0 \rangle - g)_{2.4a}$ ($(V_N \oplus N^+ - N\langle 01\bar{1}0 \rangle - g)_{1.4a}$) and $\text{Frenkel}_{\text{Oct}}(\text{Ga})$ are thermodynamically
620 ically stable rather than dissociating into two isolated point defects as predicted by the DFT
621 calculations and SW potential with respect to the interaction between them.

622 V. SUMMARY

623 In summary, the Stillinger-Weber potential is modified for GaN with the explicit consid-
624 eration of N-Ga-N (N-N-Ga) and Ga-Ga-N (Ga-N-Ga) triplets besides the regular N-Ga-Ga
625 (Ga-N-N), N-N-N and Ga-Ga-Ga terms. The fundamental properties of wurtzite GaN in-
626 cluding the lattice parameters, elastic constants and lattice dynamics are reproduced by the
627 SW and MEAM potentials. Due to the lack of long-range (e.g. electrostatic) interaction,
628 the LO-TO splitting in the phonon dispersion could be hardly described by both potentials.
629 The investigation of inversion domain boundaries indicates that the SW potential is reli-
630 able to describe the defective structures involving N-N and Ga-Ga bonds, while the MEAM
631 potential is more transferable than the SW counterpart in the structural transformation,
632 such as the wurtzite-rocksalt phase transition where the atomic environments change ap-
633 parently. The combination of SW and MEAM potentials with DFT calculation is efficient
634 in the structural search of native point defects in GaN. Two N and two Ga split interstitials
635 are first obtained in this work that are $N^+ - N\langle 2\bar{1}\bar{1}0 \rangle$, $N^+ - \text{Ga}\langle 01\bar{1}0 \rangle$ and $\text{Ga}^+ - \text{Ga}\langle 01\bar{1}0 \rangle - g$,
636 $\text{Ga}^+ - N\langle 01\bar{1}0 \rangle$ configurations after the refinement of DFT calculations from the candidates
637 relaxed by the SW and MEAM potentials, beside reproducing those already reported N
638 (Ga) vacancies and antisites, $N^+ - N\langle 01\bar{1}0 \rangle - g$ and $N^+ - N\langle 01\bar{1}0 \rangle - e$ N split interstitials, as well
639 as Ga_{Oct} , Ga_{Tet} and $\text{Ga}^+ - \text{Ga}\langle 2\bar{1}\bar{1}0 \rangle$ configurations of Ga interstitials. However, neither of
640 SW and MEAM potentials produces simultaneously the most stable N and Ga interstitials.
641 The present SW potential correctly predicts the octahedral occupation Ga_{Oct} as the most
642 stable configuration of Ga interstitial superior to the previous version^{26,38}, while the MEAM
643 potential exactly produces $N^+ - N\langle 01\bar{1}0 \rangle - g$ to be the most stable N split interstitial. On
644 a whole, the SW potential establishes less local minima than the MEAM counterpart in
645 the potential energy landscape with respect to the configurations of interstitials, whereas
646 the structural features of point defects concerning N-N or Ga-Ga bonds relaxed by the

647 SW potential are more consistent with the results of DFT calculations than those by the
648 MEAM potential. In regard to the point defect complexes, the Schottky and Frenkel_{Oct}(Ga)
649 configurations are preserved, but Frenkel_{Oct}(N) and N_{Ga}Ga_N are unstable and convert into
650 $(V_N \oplus N^+ - N\langle 01\bar{1}0 \rangle - g)_{2.4a}$ and $(V_N \oplus N^+ - N\langle 01\bar{1}0 \rangle - g)_{1.4a}$ point defect complexes in the DFT
651 calculations, respectively. In general, three methods consistently demonstrate that Schot-
652 tky is the most stable followed by $V_N \oplus N^+ - N\langle 01\bar{1}0 \rangle - g$ (either $(V_N \oplus N^+ - N\langle 01\bar{1}0 \rangle - g)_{2.4a}$ or
653 $(V_N \oplus N^+ - N\langle 01\bar{1}0 \rangle - g)_{1.4a}$) in the studied point defect complexes, and Frenkel_{Oct}(Ga) has the
654 highest formation energy. Furthermore, the formation energies calculated by DFT and SW
655 potential indicate that all of the point defect complexes are energetically favorable rather
656 than dissociating into two isolated point defects. But the MEAM potential wrongly pre-
657 dicts that the dissociation of Schottky and $V_N \oplus N^+ - N\langle 01\bar{1}0 \rangle - g$ point defect complexes is
658 exothermic. This work suggests that the present version of SW potential is applicable to
659 simulate the dynamical properties of defective GaN on a large scale with a caution spe-
660 cial to N interstitials. The mutual complementation with the MEAM potential would be a
661 practical way for getting more accurate results. Finally, the newly explored N and Ga split
662 interstitials could be useful in the study of diffusion pathways of point defects in GaN as
663 possible intermediate states, and the identification of point defects in Al(In)N alloys.

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748 ⁴⁸A directed grid search algorithm is used, where the guessed value and the variation range
749 for each parameter are set initially, then the parameter is changed step by step as far as
750 the calculated properties approach the selected references, otherwise the next parameter is
751 set to be optimized similarly. This process is repeated until no better results are obtained.

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794 TABLE I. Parameters of Stillinger-Weber potential for GaN in the format of i - j - k triplet, where
795 i is the central atom. The units of ϵ and σ are eV and Å, respectively. The parameters $\gamma = 1.2$,
796 $p = 4$ and $q = 0$ are preserved for all triplets.

Parameters	ϵ	σ	a	λ	A	B
N-Ga-Ga, Ga-N-N	2.2645	1.704	1.8	33.0	7.41825	0.69375
N-N-N	1.7368	1.400	1.8	13.0	4.41825	0.39375
Ga-Ga-Ga	1.4050	2.060	1.6	10.0	8.41825	1.09375
N-Ga-N, N-N-Ga	1.9832	1.400	1.8	13.0	5.72501	0.52265
Ga-Ga-N, Ga-N-Ga	1.7837	2.060	1.6	10.0	7.90245	0.87108

797

798 TABLE II. Lattice parameters a (Å), c (Å) and internal structural parameter u , as well as elastic
799 constants C_{ij} (GPa) and bulk modulus B (GPa) of wurtzite GaN calculated by the SW and MEAM
800 potentials in comparison with the experimental data.

GaN	Expt.	SW	MEAM
Lattice parameters			
a	3.189 ^a	3.190	3.168
c	5.186 ^a	5.210	5.173
c/a	1.626 ^a	1.633	1.633
u	0.377 ^a	0.375	0.375
Elastic parameters			
C_{11}	359.7 ^b	364.7	350.6
C_{33}	391.8 ^b	380.6	397.3
C_{44}	99.6 ^b	107.3	93.0
C_{12}	129.9 ^b	129.8	148.1
C_{13}	104.6 ^b	113.9	101.4
C_{66}	114.9 ^b	117.4	101.3
B	198.8 ^b	202.8	200.0

^a High-resolution x-ray diffraction⁵⁸.

^b ATAR experiment⁵⁹.

801

802 TABLE III. Phonon frequencies (0.1 K) at some high-symmetry k points of wurtzite GaN calculated
803 by the SW and MEAM potentials in comparison with IXS (300 K)⁶⁶ and Raman scattering (6K)
804 experiments⁶⁷ and DFPT calculations⁶⁶. All frequencies are given in THz.

k points	Mode	IXS ⁶⁶	Raman ⁶⁷	DFPT ⁶⁶	SW	MEAM
Γ	E1(LO)		22.3	22.6	25.8	26.4
Γ	A1(LO)	21.9	22.1	22.5	25.7	26.2
Γ	B1	20.7		21.6	25.5	26.0
Γ	E2		17.1	17.4	24.9	25.9
Γ	E1(TO)		16.8	17.1	24.4	25.3
Γ	A1(TO)		16.0	16.3	23.6	24.8
Γ	B1(low)	9.9		10.2	10.5	10.3
Γ	E2(low)		4.3	4.3	5.4	4.8
A	LO	21.3		22.0	25.2	26.1
A	LA	6.9		7.2	7.1	7.1
M	A1(TO)	17.3		18.0	22.9	21.4
M	E2	7.1		7.2	8.7	7.5
M	E2(low)	5.8		6.0	7.5	6.7
M	TA _z	5.5		5.6	6.4	5.6
M	TA _x	4.1		4.2	5.5	4.8
K	TO	18.4		18.8	22.0	20.3
K	TA	6.4		6.5	7.6	6.6

805

806 TABLE IV. Physical properties of IDB structures calculated by the SW and MEAM potentials in
807 comparison with DFT calculations⁵⁰. (Note: values listed are the formation energy E_{form} (meV/Å),
808 bond length (Å) of N-N (R_{NN}) and Ga-Ga (R_{GaGa}) bonds in IDB^H, as well as the averaged bond
809 length (Å) of N-Ga bonds ($R_{4\text{-fold}}$) in 4-fold atomic rings of IDB*.)

Properties DFT ⁵⁰ SW MEAM			
IDB ^H			
E_{form}	167	164	161
R_{NN}	1.51	1.57	1.19
R_{GaGa}	2.28	2.33	2.05
IDB*			
E_{form}	25	60	19
$R_{4\text{-fold}}$	1.94	2.00	1.95

810

811 TABLE V. Relative formation energies $\Delta\Omega$ (eV) of N and Ga interstitials calculated by the SW
812 and MEAM potentials along with the DFT results. $N^+-N\langle 01\bar{1}0\rangle$ -g and Ga_{Oct} configurations are
813 taken as the reference structures for N and Ga interstitials, respectively.

Configurations	SW	MEAM	DFT
N interstitials			
$N^+-N\langle 01\bar{1}0\rangle$ -g	0	0	0
$N^+-N\langle 2\bar{1}\bar{1}0\rangle$	0.310	3.363	0.407
$N^+-N\langle 01\bar{1}0\rangle$ -e	0.117	0.864	0.447
$N^+-Ga\langle 01\bar{1}0\rangle$		6.109	3.083
N_{Oct}	-0.913	4.091	
N_{Tet}		2.750	
Ga interstitials			
Ga_{Oct}	0	0	0
Ga_{Tet}	2.402		0.237
$Ga^+-Ga\langle 01\bar{1}0\rangle$ -g		0.423	0.281
$Ga^+-Ga\langle 01\bar{1}0\rangle$ -e	0.105	-0.513	
$Ga^+-N\langle 01\bar{1}0\rangle$		0.363	0.391
$Ga^+-N\langle 01\bar{1}0\rangle$ -o		0.373	
$Ga^+-Ga\langle 2\bar{1}\bar{1}0\rangle$	0.484	0.899	1.407

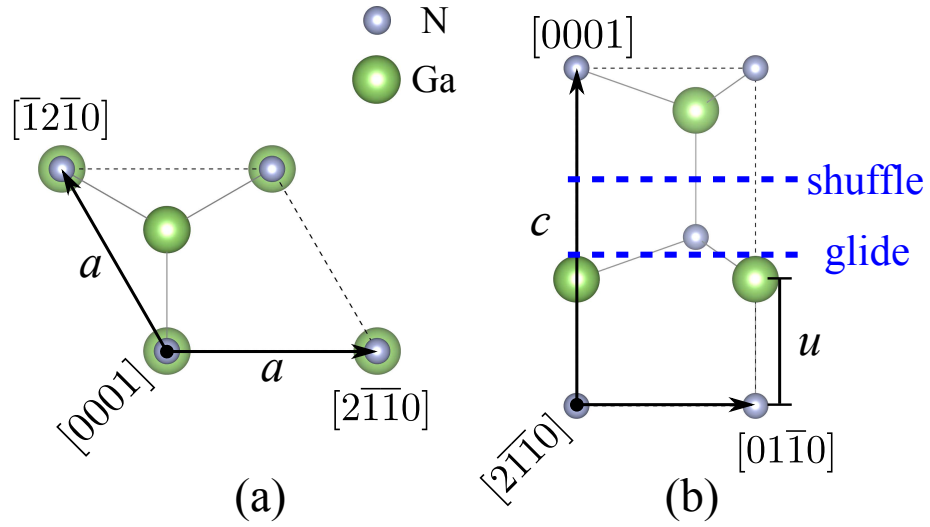
814

815 TABLE VI. Formation energies Ω (eV) (Eq. 4) of point defect complexes and paired point defects
 816 calculated by DFT as well as the SW and MEAM potentials, respectively.

Configurations	DFT	SW	MEAM
Point defect complexes			
Schottky	6.669	6.530	4.148
Frenkel _{Oct} (N)		8.680 ^a	9.443 ^a
(V _N ⊕ N ⁺ -N⟨011̄0⟩-g) _{2.4a}	7.328	9.848 ^b	5.519 ^b
N _{Ga} Ga _N		8.605 ^a	13.076 ^a
(V _N ⊕ N ⁺ -N⟨011̄0⟩-g) _{1.4a}	7.497	7.207 ^b	5.567 ^b
Frenkel _{Oct} (Ga)	10.071	10.456	6.072
Paired point defects			
V _N + V _{Ga}	10.213	8.706	4.114
V _N + N ⁺ -N⟨011̄0⟩-g	8.044	9.864	5.495
N _{Ga} + Ga _N	13.900	13.211	13.152
V _{Ga} + Ga _{Oct}	14.950	10.481	6.136

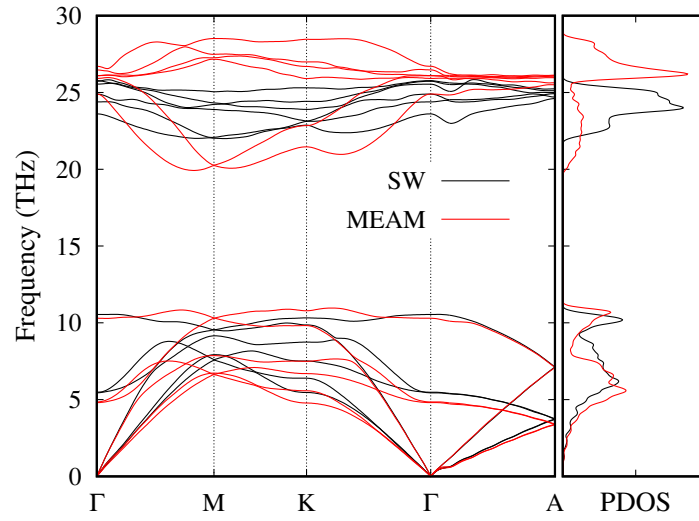
^a Preserved atomic structures.

^b DFT-relaxed structure as initial configuration.



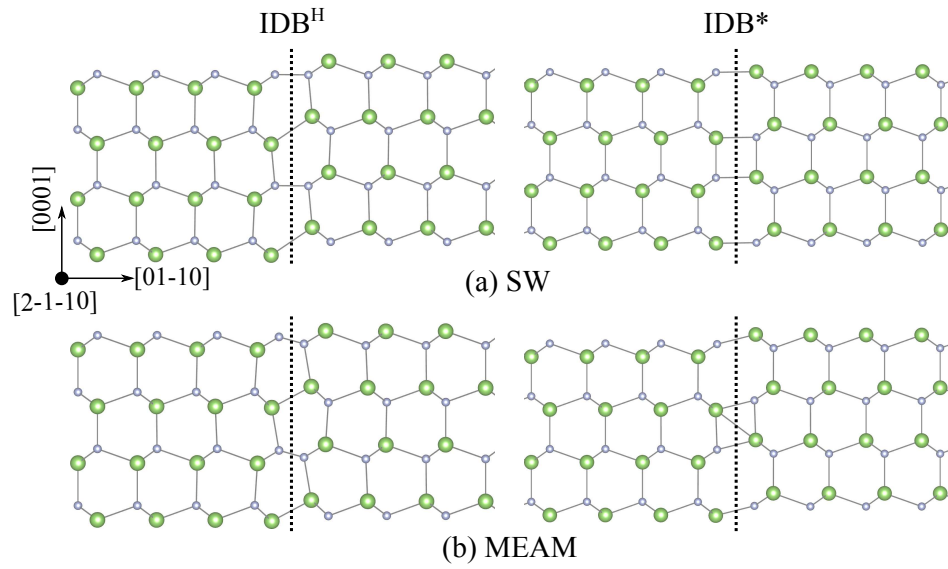
818

819 FIG. 1. (Color online) Sketch of the primitive cell of wurtzite GaN. (a) $[0001]$ projection. (b)
 820 $[2\bar{1}\bar{1}0]$ projection. Small gray and large green balls represent N and Ga atoms, respectively. a ,
 821 c and u are the lattice constants and internal structural parameter, respectively. The glide and
 822 shuffle spaces along the $[0001]$ plane are labeled out. VESTA⁷⁶ is used for visualization.



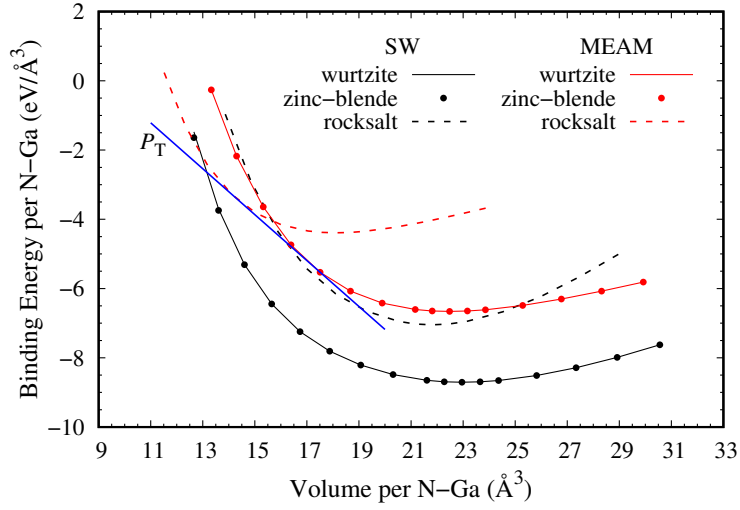
823

824 FIG. 2. (Color online) Phonon dispersion relationship of wurtzite GaN along the high symmetry
 825 lines of Brillouin zone at 0.1 K calculated by the SW and MEAM potentials, respectively.



826

827 FIG. 3. (Color online) Atomic structures of IDB^H (Left) and IDB* (Right) in wurtzite GaN
 828 calculated by (a) SW and (b) MEAM potentials, respectively. The boundaries are indicated by
 829 the dashed lines. The atomic representations are the same as in Fig. 1.



830

831 FIG. 4. (Color online) Volume dependence of binding energy in wurtzite, zinc-blende and rock-
 832 salt lattices per formula unit (Ga-N) calculated by the SW (black) and MEAM (red) potentials,
 833 respectively. P_T is the equilibrium transition pressure of wurtzite-rocksalt phase transition, which
 834 is estimated to be 106 GPa by the MEAM potential.

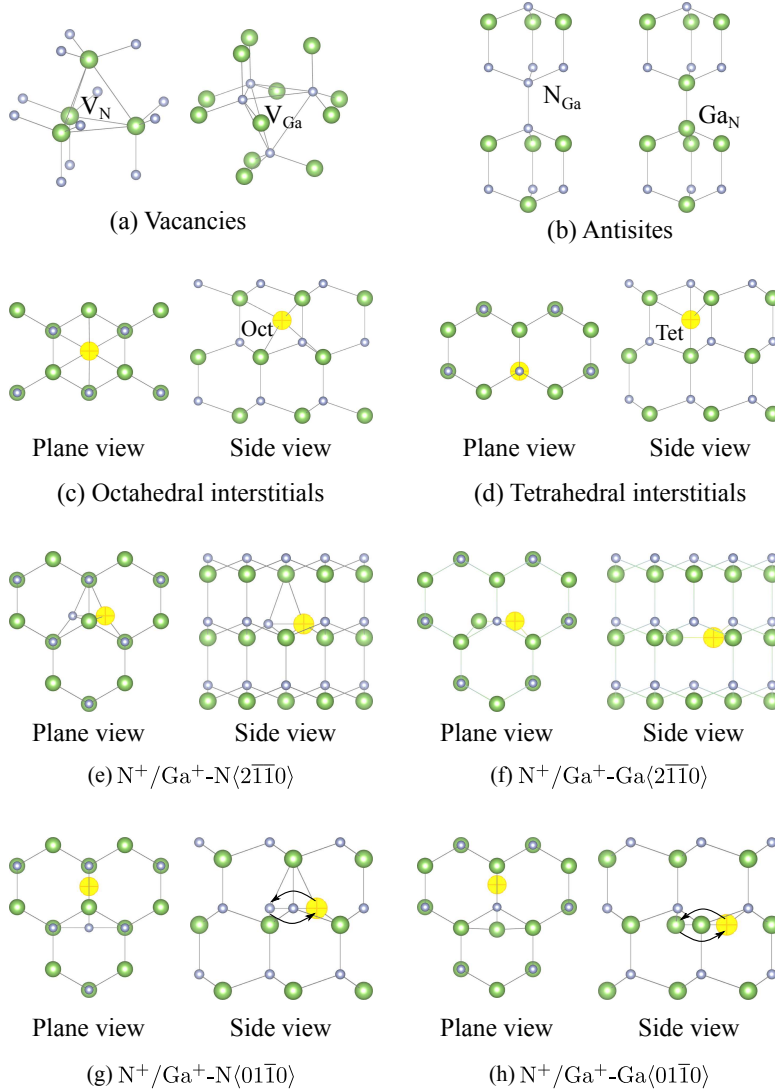


FIG. 5. (Color online) Illustrations of initial atomic configurations of native point defects in wurtzite GaN. (a) Vacancies, V_N and V_{Ga} . (b) Antisites, N_{Ga} and Ga_N . (c) Octahedral interstitials, N_{Oct} and Ga_{Oct} . (d) Tetrahedral interstitials, N_{Tet} and Ga_{Tet} . (e) $\langle 2\bar{1}\bar{1}0 \rangle$ -oriented split interstitials formed with a host N atom, $N^+-N\langle 2\bar{1}\bar{1}0 \rangle$ and $Ga^+-N\langle 2\bar{1}\bar{1}0 \rangle$. (f) $\langle 2\bar{1}\bar{1}0 \rangle$ -oriented split interstitials formed with a host Ga atom, $N^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$ and $Ga^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$. (g) $\langle 01\bar{1}0 \rangle$ -oriented split interstitials formed with a host N atom, $N^+-N\langle 01\bar{1}0 \rangle$ and $Ga^+-N\langle 01\bar{1}0 \rangle$. (h) $\langle 01\bar{1}0 \rangle$ -oriented split interstitials formed with a host Ga atom, $Ga^+-Ga\langle 01\bar{1}0 \rangle$ and $N^+-Ga\langle 01\bar{1}0 \rangle$. The representations of atomic structures are the same as in Fig. 1. Large yellow balls in (c)-(h) label out the occupation sites for N/Ga atoms to form the N^+/Ga^+ interstitials. Arrows in (g) and (h) represent the exchange of N and Ga sites to form the opposite $\langle 01\bar{1}0 \rangle$ -oriented N-Ga dumbbells in the construction of $Ga^+-N\langle 01\bar{1}0 \rangle$ -o and $N^+-Ga\langle 01\bar{1}0 \rangle$ -o split interstitials, respectively.

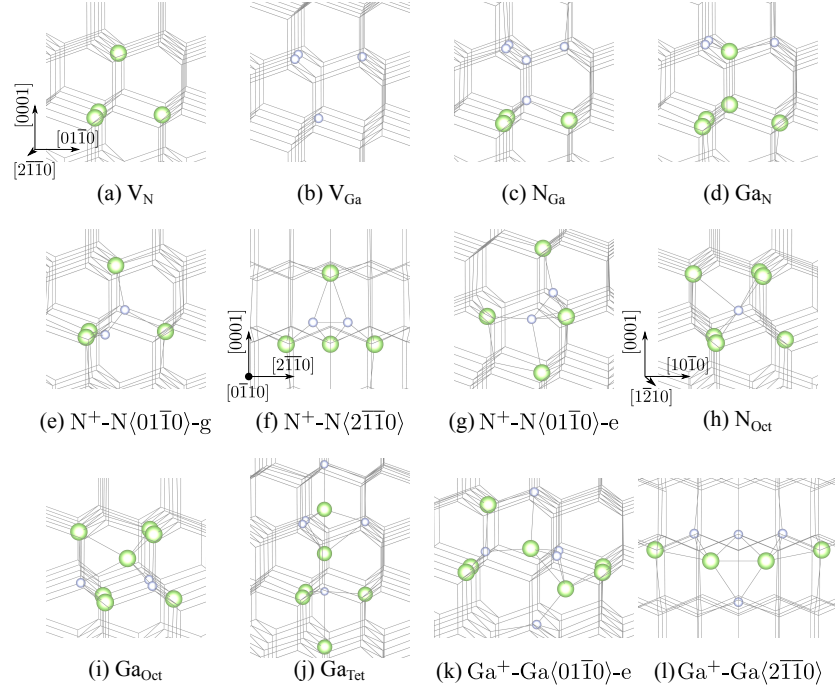


FIG. 6. (Color online) Perspective view of atomic structures of native point defects in wurtzite GaN relaxed by the SW potential. (a) V_N . (b) V_{Ga} . (c) N_{Ga} . (d) Ga_N . (e) $N^+-N\langle 01\bar{1}0 \rangle$ -g. (f) $N^+-N\langle 2\bar{1}\bar{1}0 \rangle$. (g) $N^+-N\langle 01\bar{1}0 \rangle$ -e. (h) N_{Oct} . (i) Ga_{Oct} . (j) Ga_{Tet} . (k) $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -e. (l) $Ga^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$. Small gray and large green balls represent the neighboring N and Ga atoms around the point defects, respectively. Other atoms and bonds in bulk GaN are simplified by the wire frames. The coordinate system illustrated in (a) is used by default, except the cases in (f) and (l) where the coordinate system is illustrated in (f), and the cases in (h) and (i) whose coordinate system is illustrated in (h).

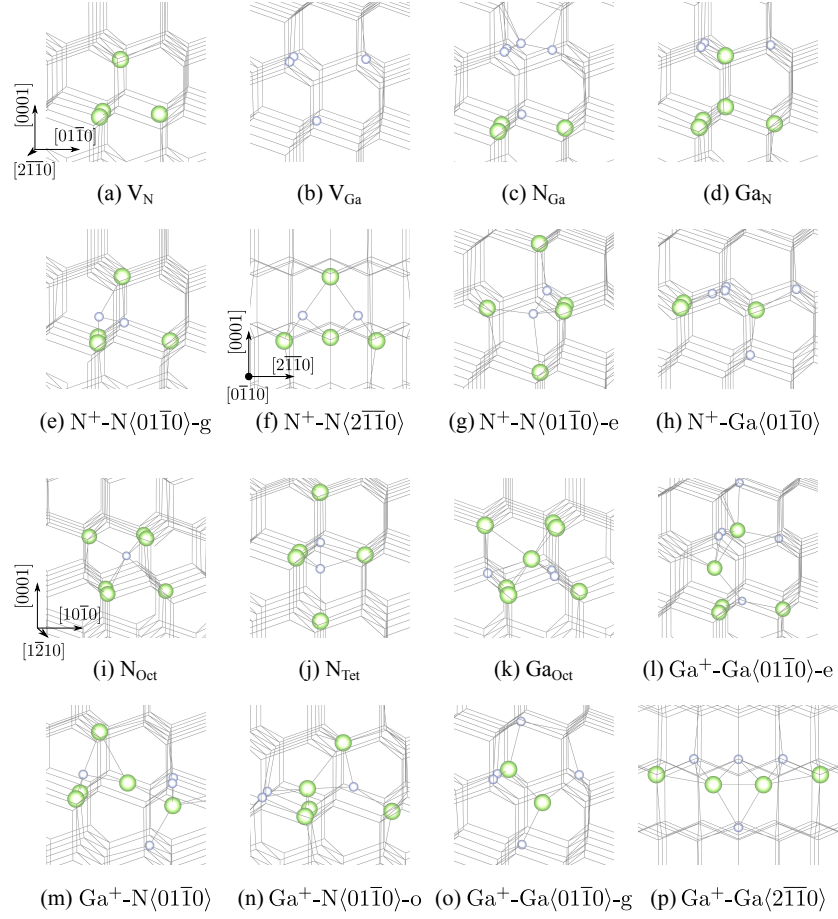


FIG. 7. (Color online) Perspective view of atomic structures of native point defects in wurtzite GaN relaxed by the MEAM potential. (a) V_N . (b) V_{Ga} . (c) N_{Ga} . (d) Ga_N . (e) $N^+-N\langle 01\bar{1}0 \rangle\text{-g}$. (f) $N^+-N\langle 2\bar{1}\bar{1}0 \rangle$. (g) $N^+-N\langle 01\bar{1}0 \rangle\text{-e}$. (h) $N^+-Ga\langle 01\bar{1}0 \rangle$. (i) N_{Oct} . (j) N_{Tet} . (k) Ga_{Oct} . (l) $Ga^+-Ga\langle 01\bar{1}0 \rangle\text{-e}$. (m) $Ga^+-N\langle 01\bar{1}0 \rangle$. (n) $Ga^+-N\langle 01\bar{1}0 \rangle\text{-o}$. (o) $Ga^+-Ga\langle 01\bar{1}0 \rangle\text{-g}$. (p) $Ga^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$. The representations of atomic structures are the same as in Fig. 6. The coordinate system illustrated in (a) is used by default, except the cases in (f) and (p) where the coordinate system is illustrated in (f), and the cases in (i) and (k) where the coordinate system is illustrated in (i).

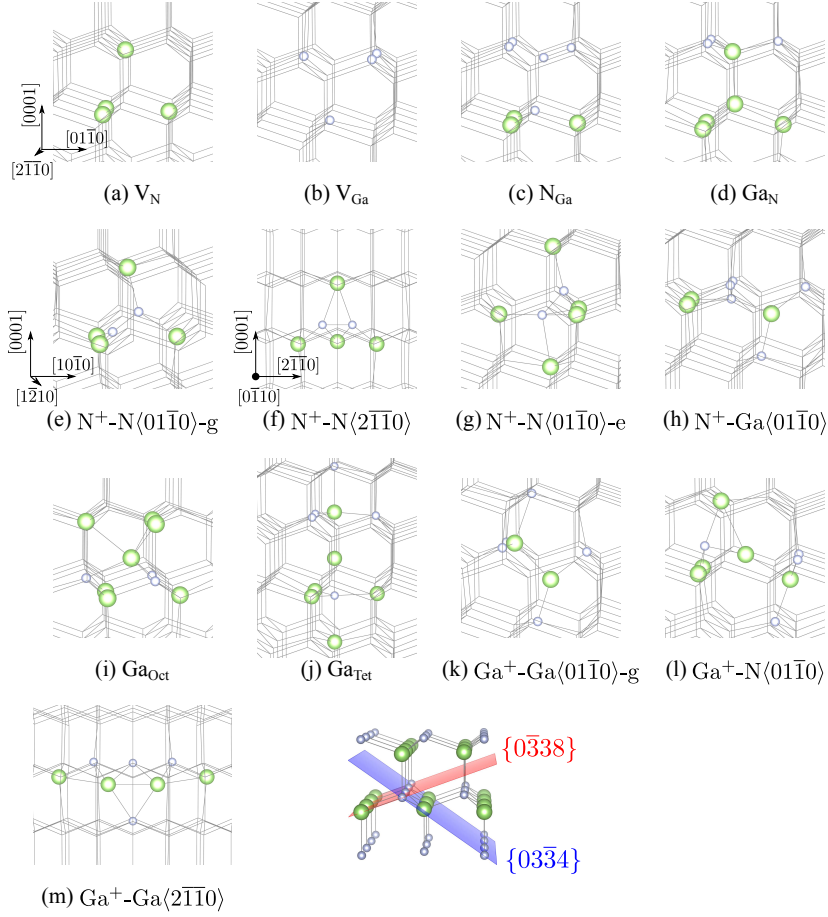


FIG. 8. (Color online) Perspective view of atomic structures of native point defects in wurtzite GaN relaxed by DFT calculations. (a) V_N . (b) V_{Ga} . (c) N_{Ga} . (d) Ga_N . (e) $N^+-N\langle 01\bar{1}0 \rangle$ -g. (f) $N^+-N\langle 2\bar{1}\bar{1}0 \rangle$. (g) $N^+-N\langle 01\bar{1}0 \rangle$ -e. (h) $N^+-Ga\langle 01\bar{1}0 \rangle$. (i) Ga_{Oct} . (j) Ga_{Tet} . (k) $Ga^+-Ga\langle 01\bar{1}0 \rangle$ -g. (l) $Ga^+-N\langle 01\bar{1}0 \rangle$. (m) $Ga^+-Ga\langle 2\bar{1}\bar{1}0 \rangle$. The representations of atomic structures are the same as in Fig. 6. The coordinate system illustrated in (a) is used by default, except the cases in (e) and (i) where the coordinate system is illustrated in (e), and the cases in (f) and (m) where the coordinate system is illustrated in (f). Inset indicates the $\{0\bar{3}38\}$ plane (in red) where N-Ga pairs arrange in parallel, and the $\{03\bar{3}4\}$ plane (in blue) where N-Ga pairs arrange in zigzag pattern.

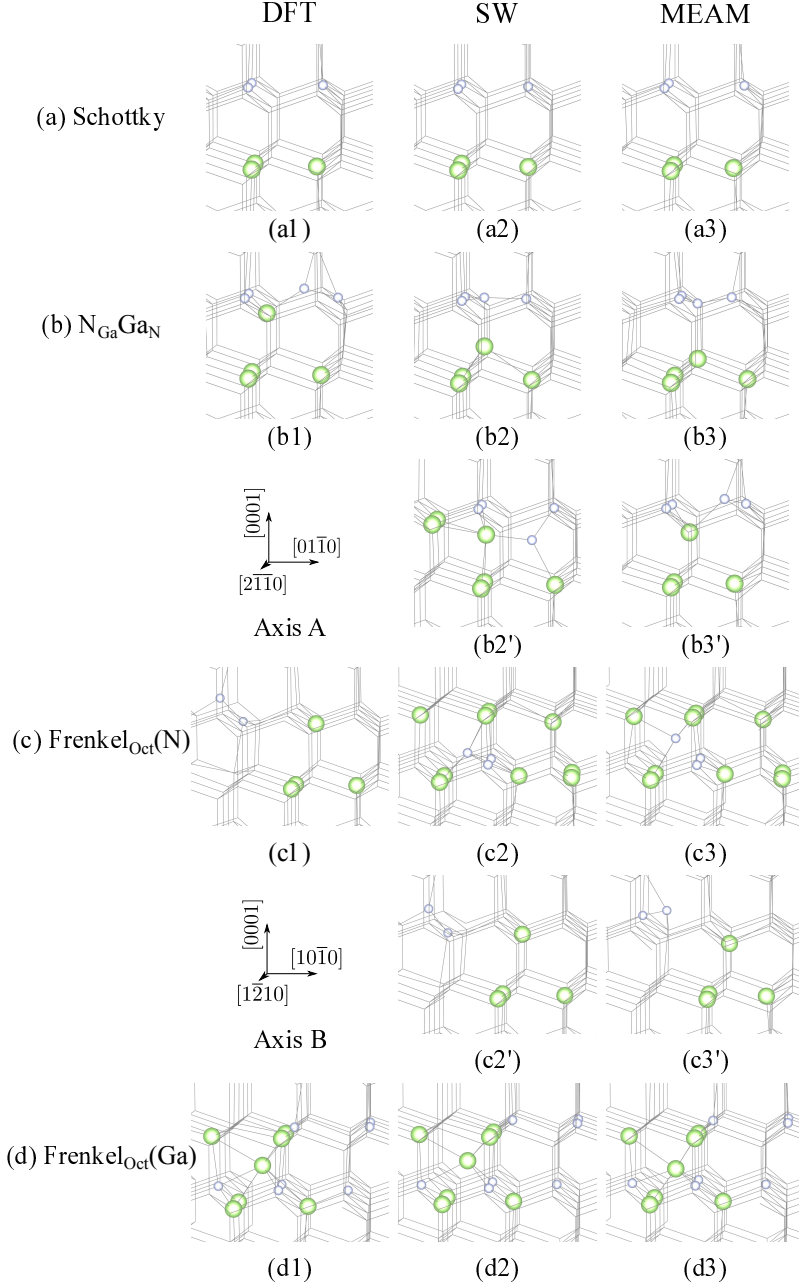
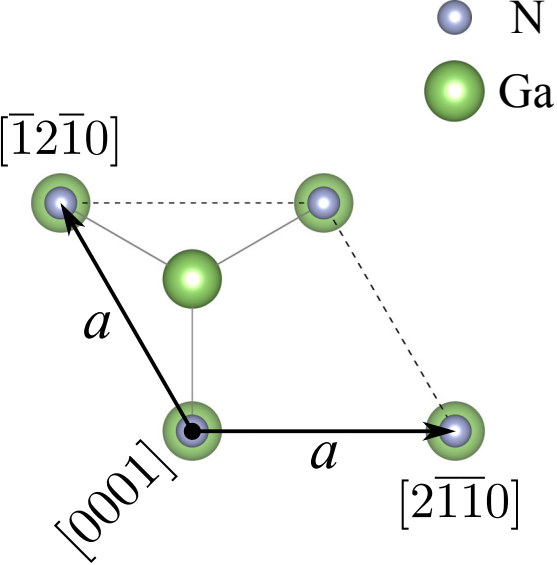
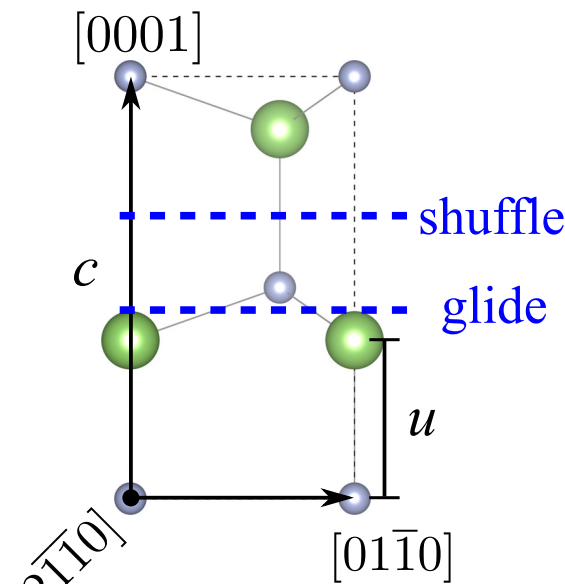


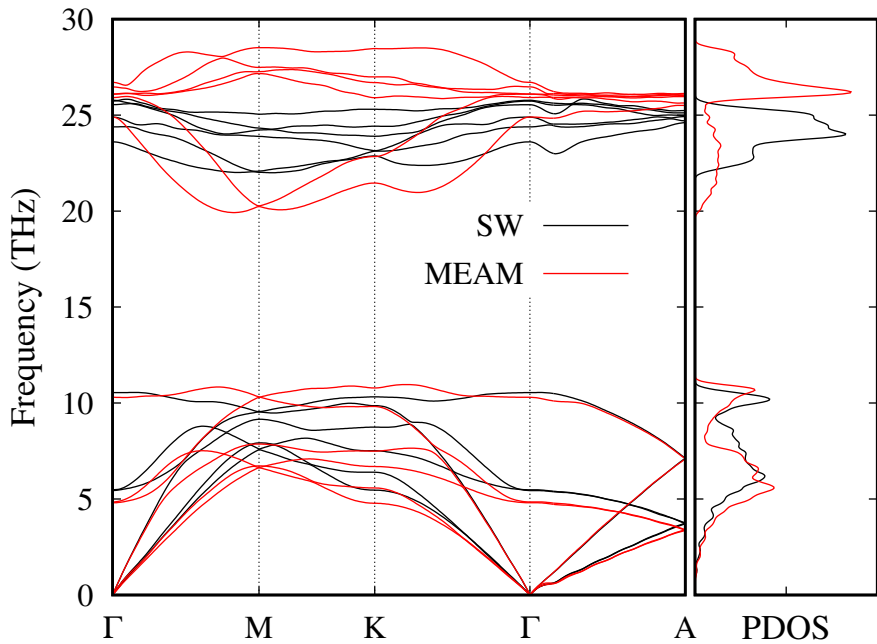
FIG. 9. (Color online) Perspective view of atomic structures of point defect complexes in wurtzite GaN relaxed by DFT (Left), SW (Middle) and MEAM (Right) potentials, respectively. (a) Schottky. (b) $N_{Ga}Ga_N$, where (b2') and (b3') are the relaxed structures by taking the DFT result in (b1) as the initial configuration. (c) $Frenkel_{Oct}(N)$, where (c2') and (c3') are the relaxed structures by taking the DFT result in (c1) as the initial configuration. And (d) $Frenkel_{Oct}(Ga)$. The representations of atomic structures are the same as in Fig. 6. The coordinate system denoted by 'Axis A' is used by default, except the cases in (c2), (c3), (d1), (d2) and (d3), which are coordinated by the axes denoted as 'Axis B'.

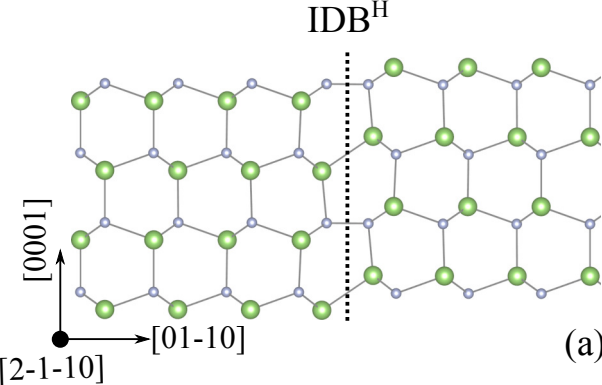


(a)

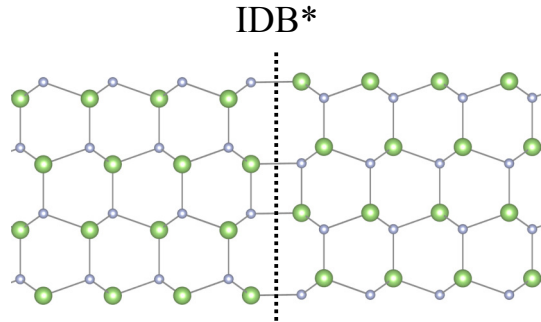


(b)

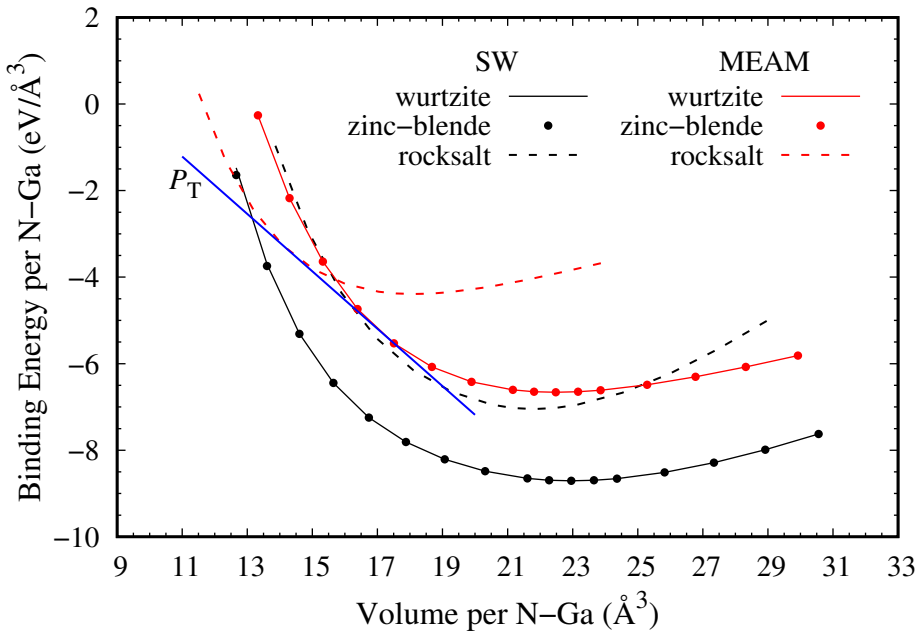


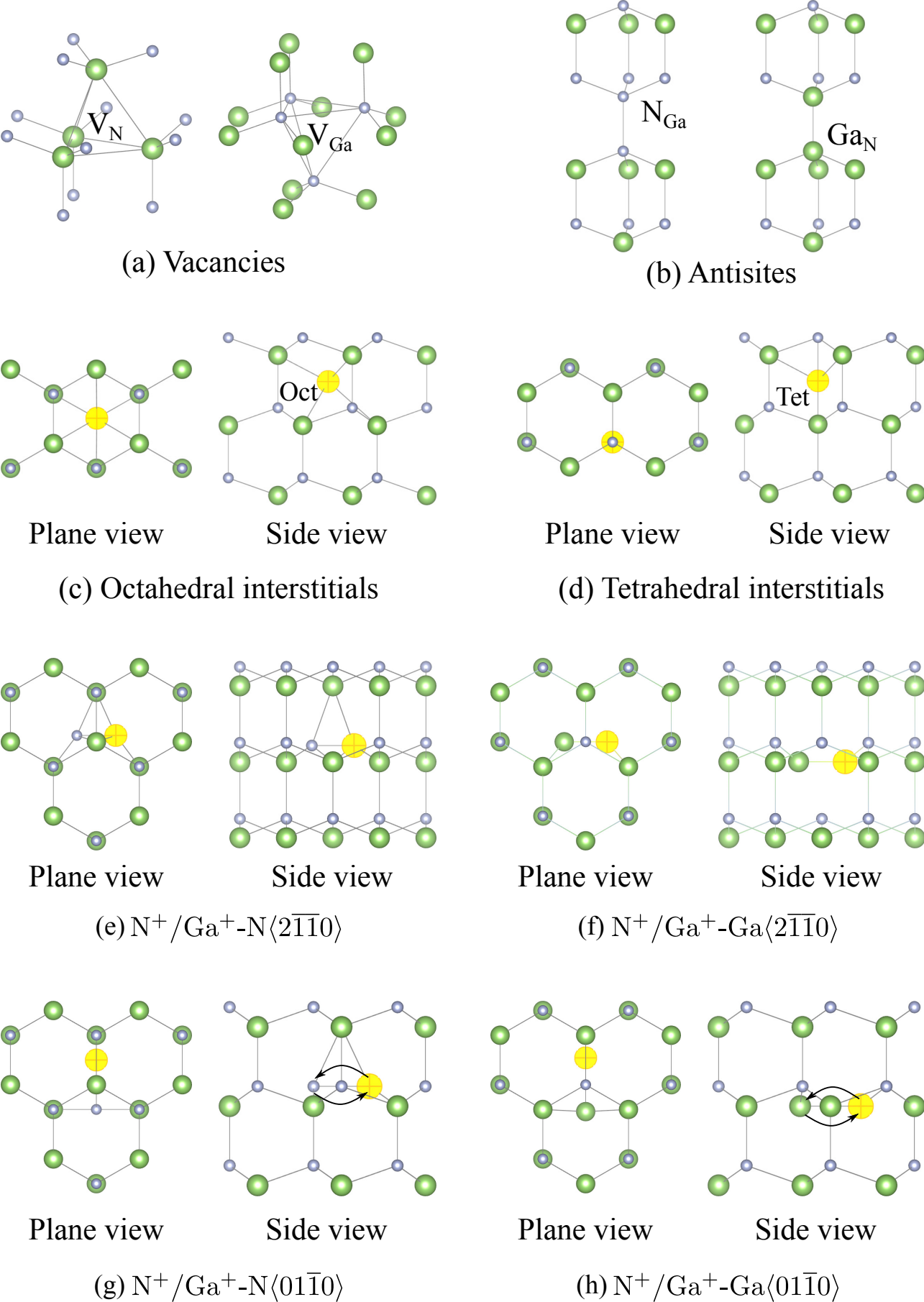


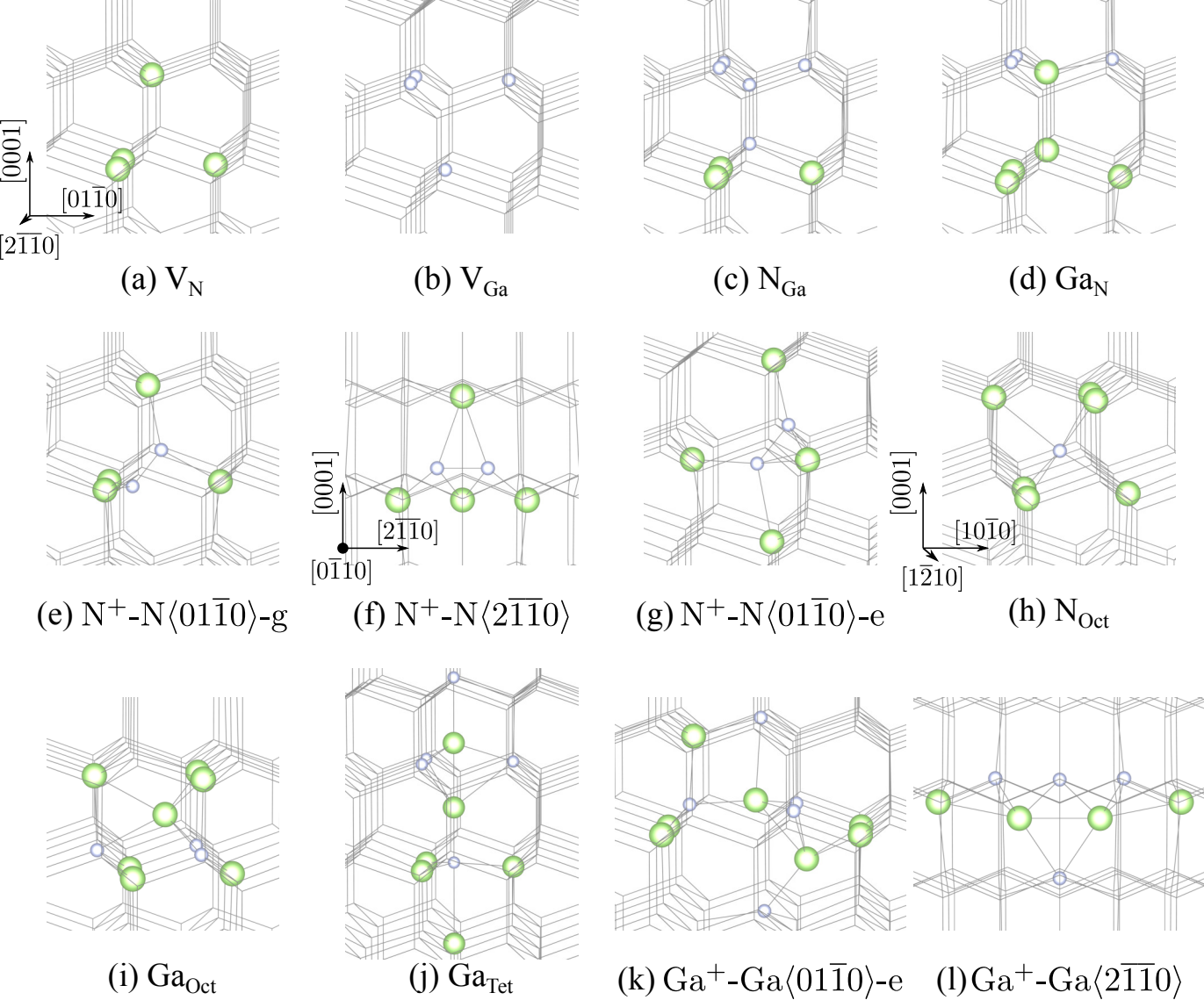
(a) SW

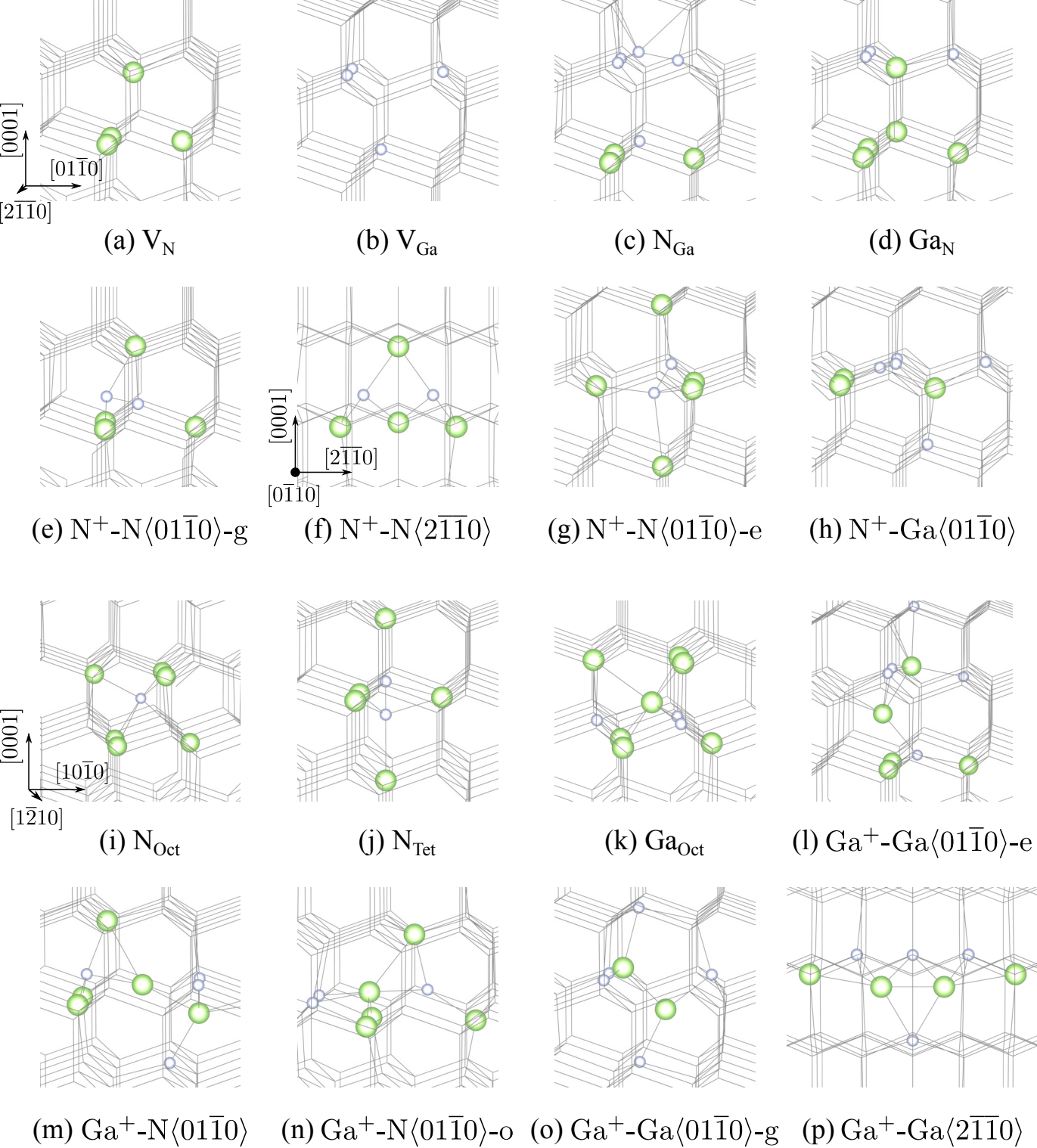


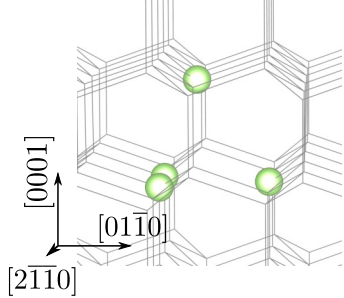
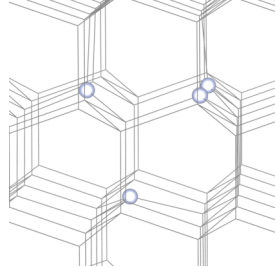
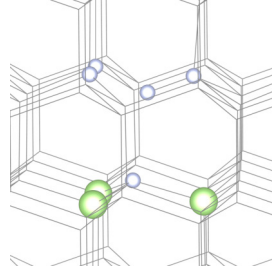
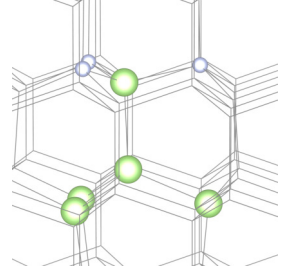
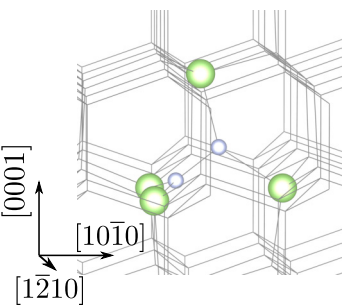
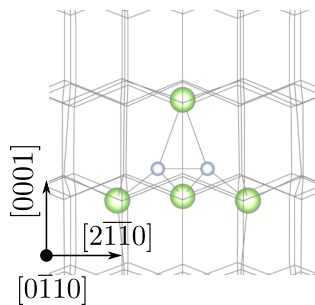
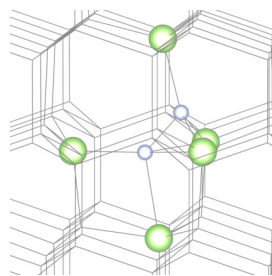
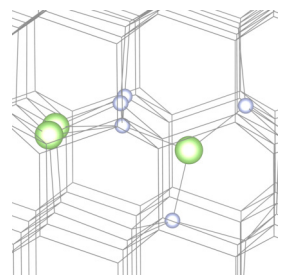
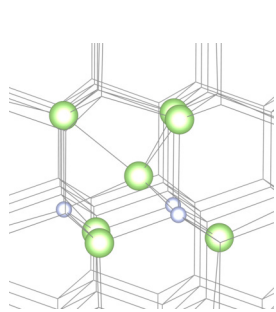
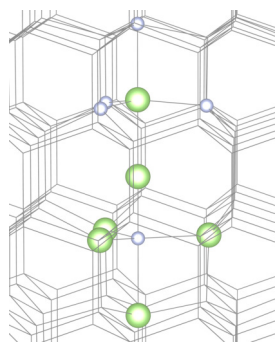
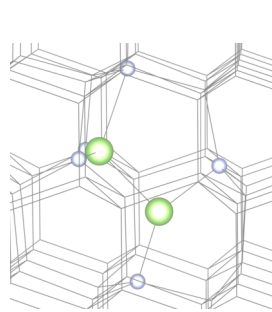
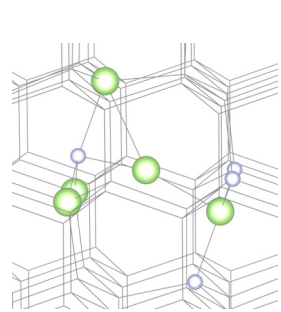
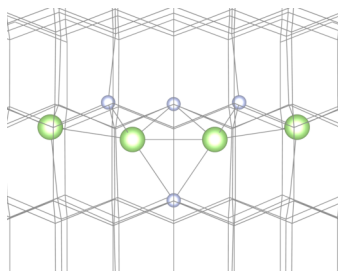
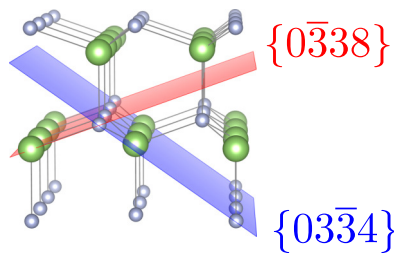
(b) MEAM









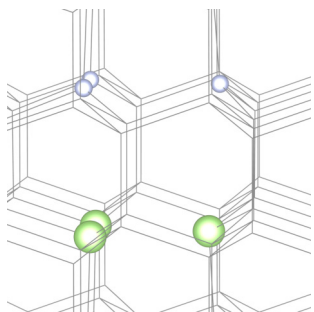
(a) V_N (b) V_{Ga} (c) N_{Ga} (d) Ga_N (e) $N^+-N\langle 01\bar{1}0\rangle-g$ (f) $N^+-N\langle 2\bar{1}\bar{1}0\rangle$ (g) $N^+-N\langle 01\bar{1}0\rangle-e$ (h) $N^+-Ga\langle 01\bar{1}0\rangle$ (i) Ga_{Oct} (j) Ga_{Tet} (k) $Ga^+-Ga\langle 01\bar{1}0\rangle-g$ (l) $Ga^+-N\langle 01\bar{1}0\rangle$ (m) $Ga^+-Ga\langle 2\bar{1}\bar{1}0\rangle$ 

DFT

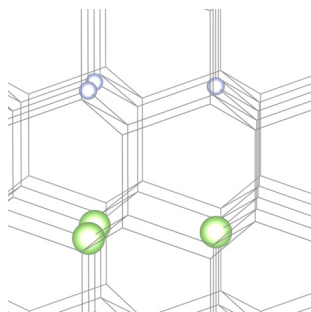
SW

MEAM

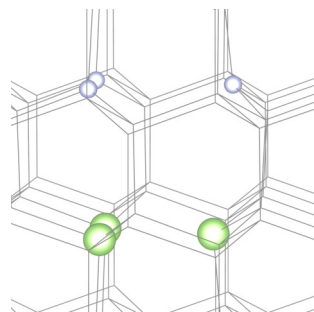
(a) Schottky



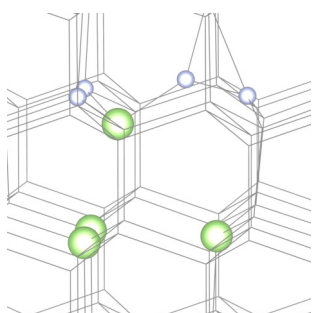
(a1)



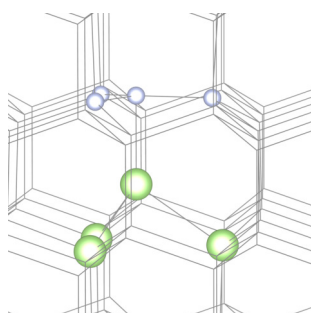
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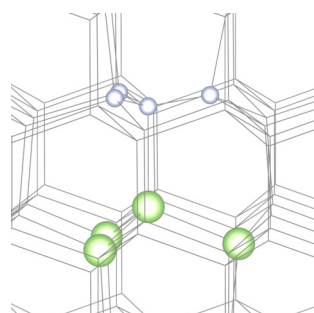
(a3)

(b) $N_{\text{Ga}}\text{Ga}_\text{N}$ 

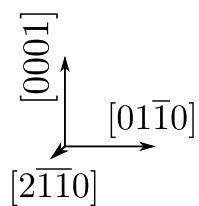
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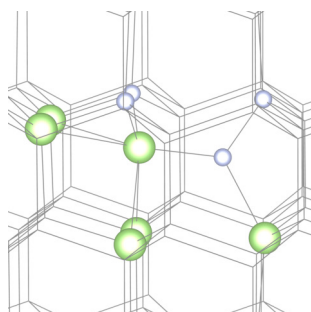
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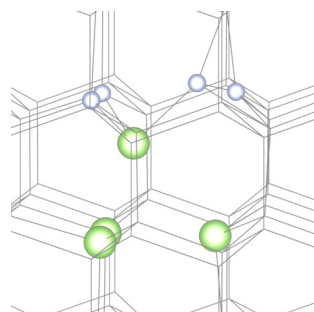
(b3)



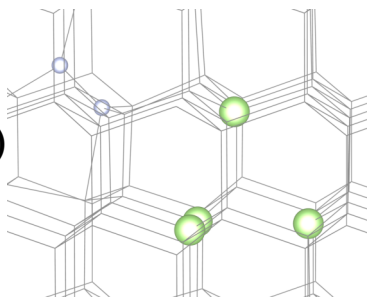
Axis A



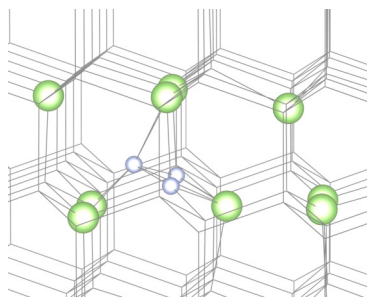
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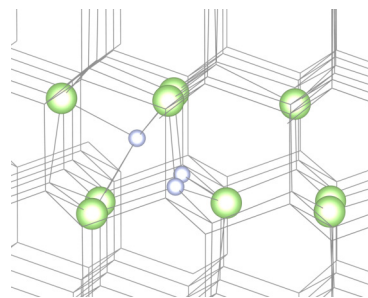
(b3')

(c) $\text{Frenkel}_{\text{Oct}}(\text{N})$ 

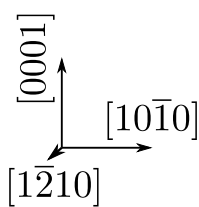
(c1)



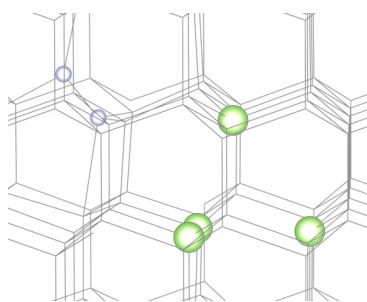
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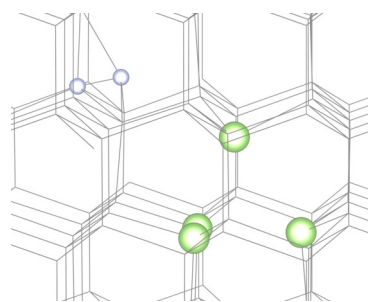
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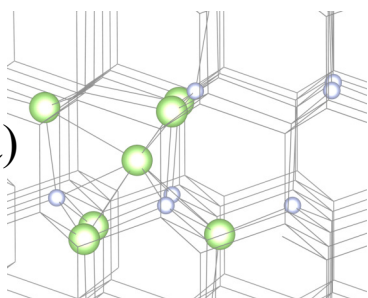
Axis B



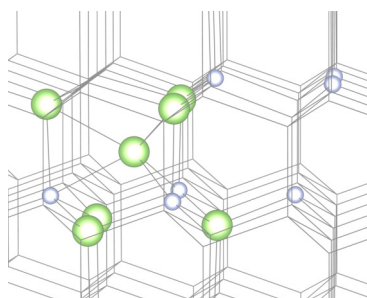
(c2')



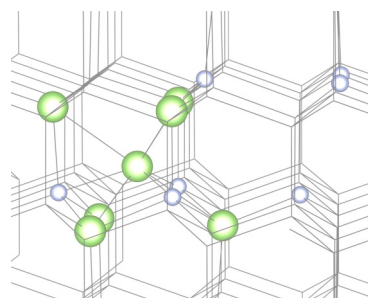
(c3')

(d) $\text{Frenkel}_{\text{Oct}}(\text{Ga})$ 

(d1)



(d2)



(d3)