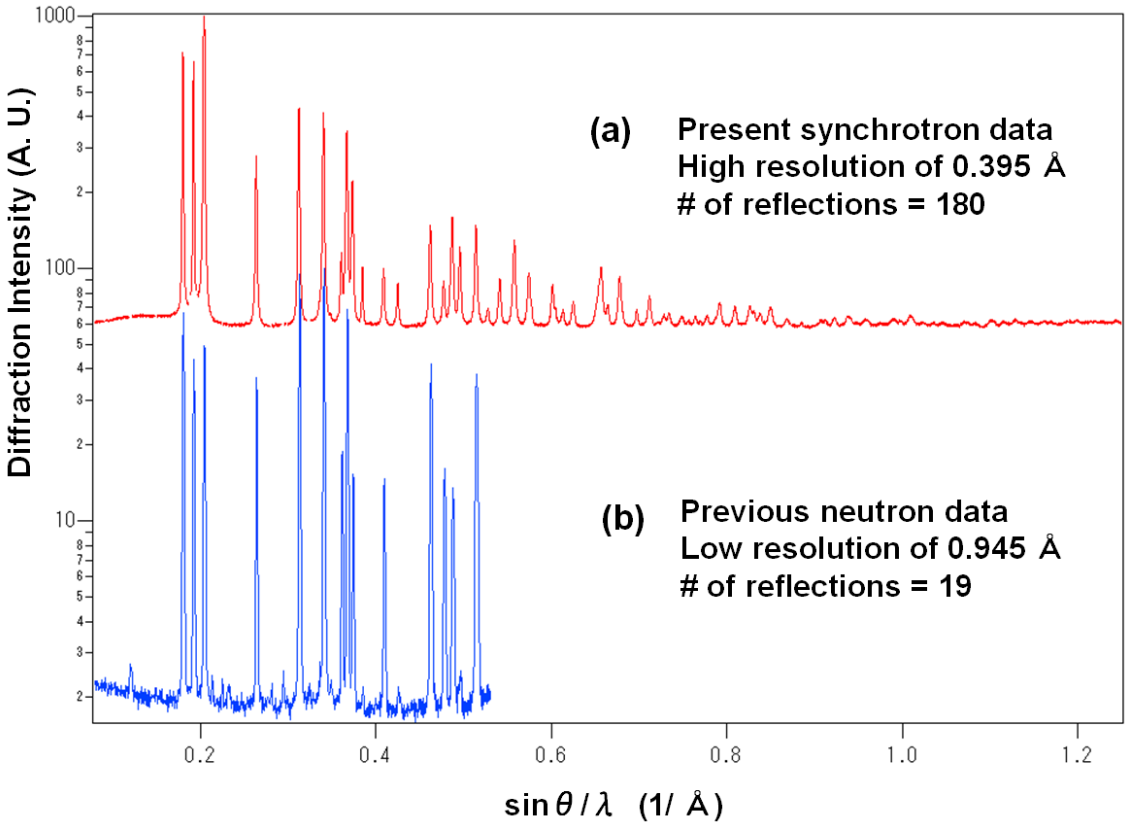

Figure S1. Comparison of (a) the present synchrotron powder diffraction data of $(Ga_{0.885}Zn_{0.115})(N_{0.885}O_{0.115})$ (Higher resolution of 0.395 Å, Wavelength of incident X-ray = 0.49942 Å) and (b) previous neutron powder diffraction data (Lower resolution of 0.945 Å, Wavelength of neutron beam = 1.82035 Å). The number of observed reflections (180) in this work is much larger than that (19) in the previous work [Ref. 1], which enables the detailed analysis of structural disorder in $(Ga_{0.885}Zn_{0.115})(N_{0.885}O_{0.115})$.

[Ref. 1] Yashima, M. *et al. Chem. Phys. Lett.* **2005**, *416*, 225-228.

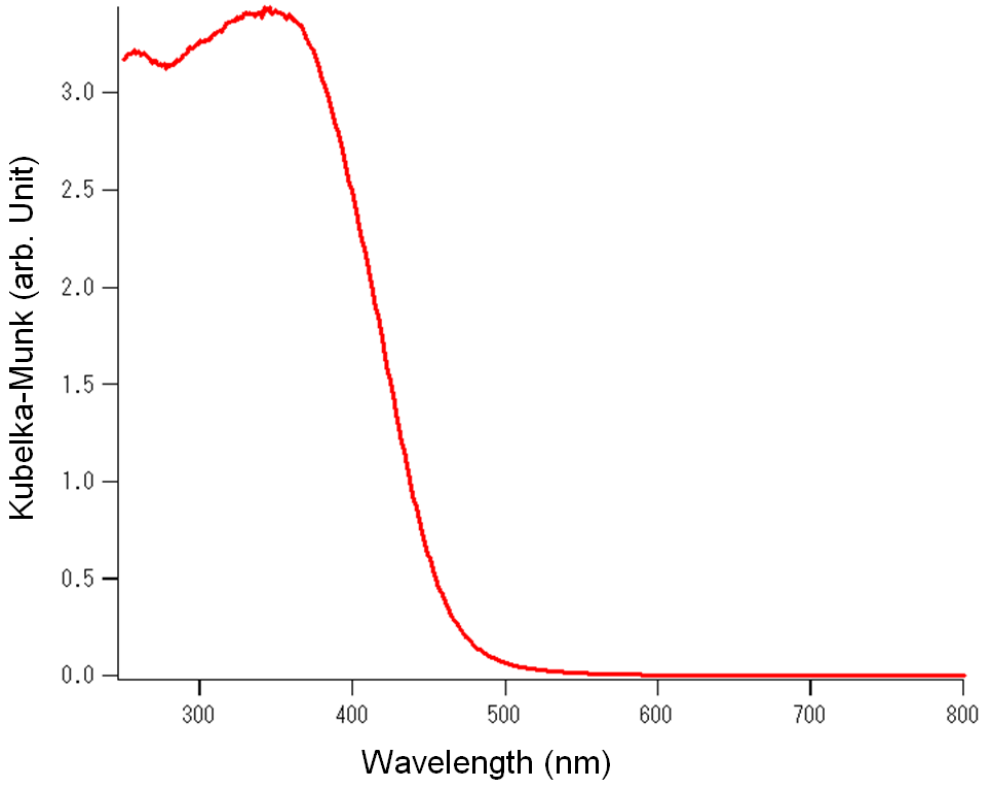

Figure S2. UV-visible diffuse reflectance spectrum of $(Ga_{0.885}Zn_{0.115})(N_{0.885}O_{0.115})$. The band gap was estimated to be 2.7 eV from this figure.

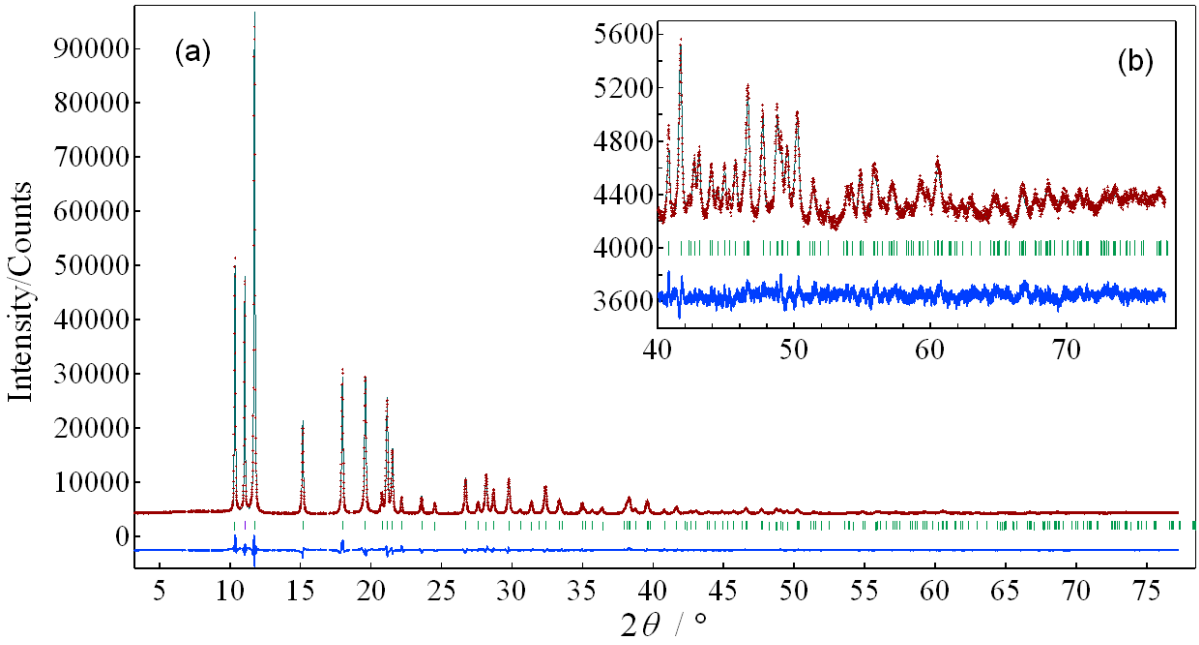

Figure S3. (a) Rietveld pattern for the non-split-atom model for synchrotron powder diffraction data of $(Ga_{0.885}Zn_{0.115})(N_{0.885}O_{0.115})$ at 305 K. Red plots denote observed data, green line denotes calculated profiles, and blue line denotes the difference. Vertical lines indicate possible Bragg peaks of the hexagonal $(Ga_{0.885}Zn_{0.115})(N_{0.885}O_{0.115})$ phase. Wavelength of incident X-ray is 0.49942 Å. Inset (b) is the enlargement of the higher angle region.

Table S1. Refined crystallographic parameters and reliability factors in the Rietveld analysis of synchrotron X-ray powder diffraction data of $(Ga_{0.885}Zn_{0.115})(N_{0.885}O_{0.115})$ (305 K). Anions were put at single 1/3, 2/3, 0 site (non-split-atom model).

Atom	Site	Occupancy	Atomic coordinates		
		g	*x*	*y*	*z*
Ga,Zn	2*b*	1.0	1/3	2/3	0.3756(4)
N	2*b*	0.885	1/3	2/3	0.0
O	2*b*	0.115	1/3	2/3	0.0

Unit cell: hexagonal $P6_3mc$, $a = b = 3.19833(3)$ Å, $c = 5.19217(5)$ Å, $\alpha = \beta = 90°$, $\gamma = 120°$, $V = 45.9966(8)$ Å^3.
Atomic displacement parameters: U(Ga) = U(Zn) = U(N) = U(O) = 0.0063(2) Å^2
$R_{wp} = 1.90$ %, GOF = 1.35, $R_I = 1.77$ %, $R_F = 1.16$ %
Wavelength = 0.49942 Å

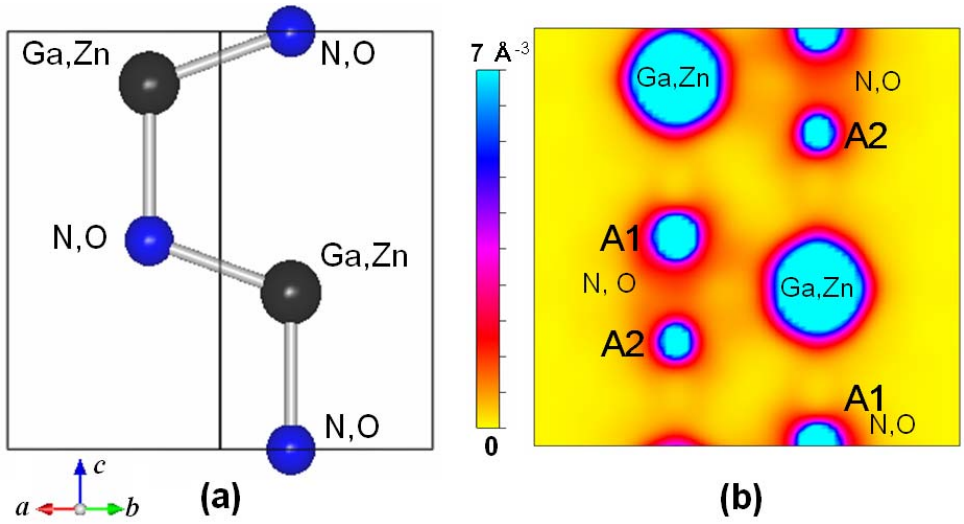

Figure S4. (a) Projection of refined crystal structure of $(Ga_{0.885}Zn_{0.115})(N_{0.885}O_{0.115})$ on the (110) plane. (b) Electron-density distribution on the (110) plane of $(Ga_{0.885}Zn_{0.115})(N_{0.885}O_{0.115})$. A1 and A2 are the split sites of anions. Structure refinement is performed with a non-split-atom model.

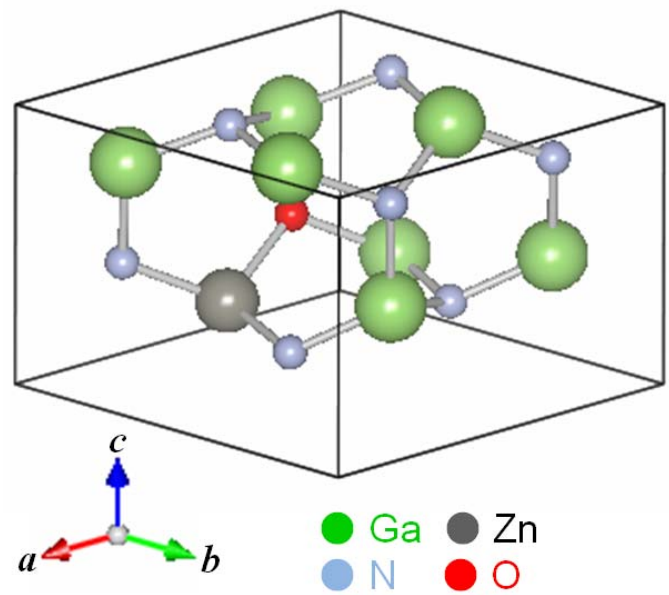

Figure S5. 2×2×1 supercell of the hexagonal GaN-ZnO phase, $(Ga_7Zn)(N_7O)$, which was optimized by the DFT calculations. There exist seven Ga, one Zn, seven N and one O atoms in the supercell. Details of the DFT calculations are described below. CIF file of this structure is shown in Table S2.

Table S2. CIF file for the optimized $(Ga_7Zn)(N_7O)$.

```
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_symmetry_space_group_name_H-M     'P1'
_symmetry_Int_Tables_number       1

_cell_length_a                   6.32426
_cell_length_b                   6.29318
_cell_length_c                   5.44533
_cell_angle_alpha                 89.98
_cell_angle_beta                 90.12
_cell_angle_gamma                120.21
_cell_volume                     187.28
_cell_formula_units_Z             1

loop_
_atom_site_label
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_symbol
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
Ga      1 a   0.15354   0.32274   0.29761 1.0
Ga      1 a   0.33806   0.17461   0.81128 1.0
N       1 a   0.15966   0.31620   0.94069 1.0
O       1 a   0.32740   0.17832   0.44317 1.0
Ga      1 a   0.15079   0.80720   0.44621 1.0
Ga      1 a   0.34172   0.67944   0.94261 1.0
N       1 a   0.15971   0.81791   0.80141 1.0
N       1 a   0.34246   0.68414   0.30518 1.0
Zn      1 a   0.67193   0.32489   0.29757 1.0
Ga      1 a   0.83807   0.17793   0.79823 1.0
N       1 a   0.65918   0.32068   0.92002 1.0
N       1 a   0.84942   0.17930   0.44517 1.0
Ga      1 a   0.66605   0.82547   0.44187 1.0
Ga      1 a   0.83575   0.67613   0.94240 1.0
N       1 a   0.65680   0.81601   0.80424 1.0
N       1 a   0.84944   0.69902   0.29697 1.0
```

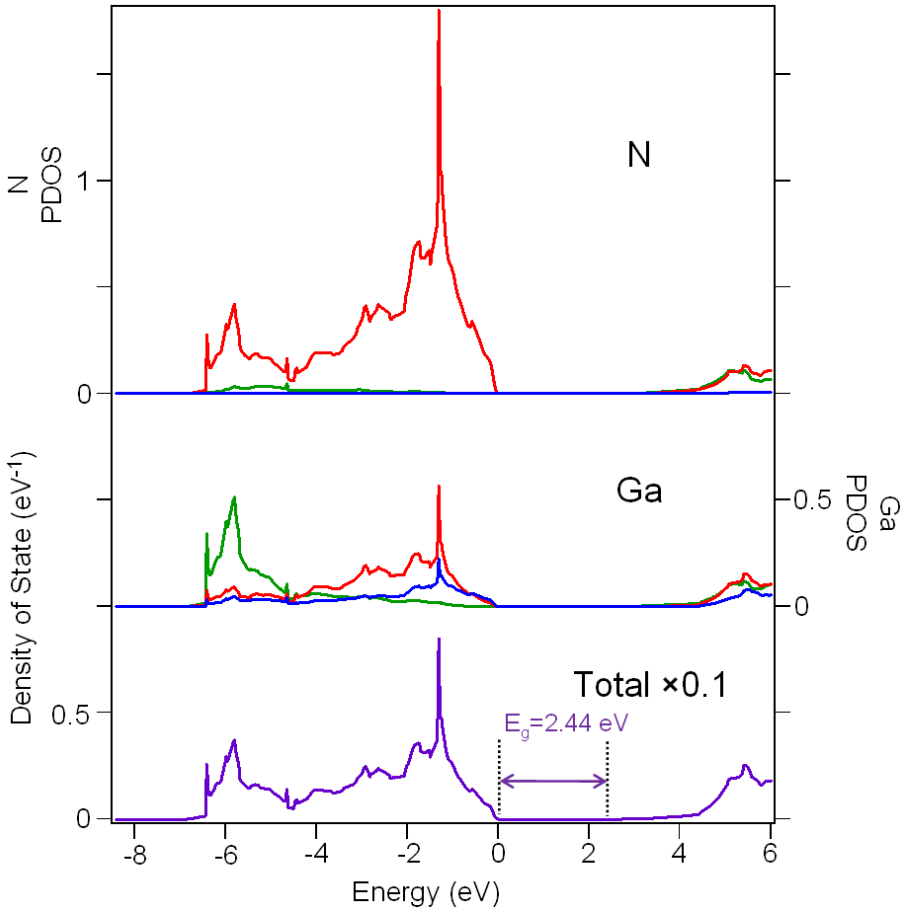

Figure S6. Total density of states (purple curve) and partial density of states (PDOS) of Ga and N atoms in GaN. Green, red and blue lines denote the s, p and d orbitals, respectively. These curves were obtained by the DFT calculations. The band gap 2.44 eV was estimated from the dispersion curve depicted in Fig. S8A.

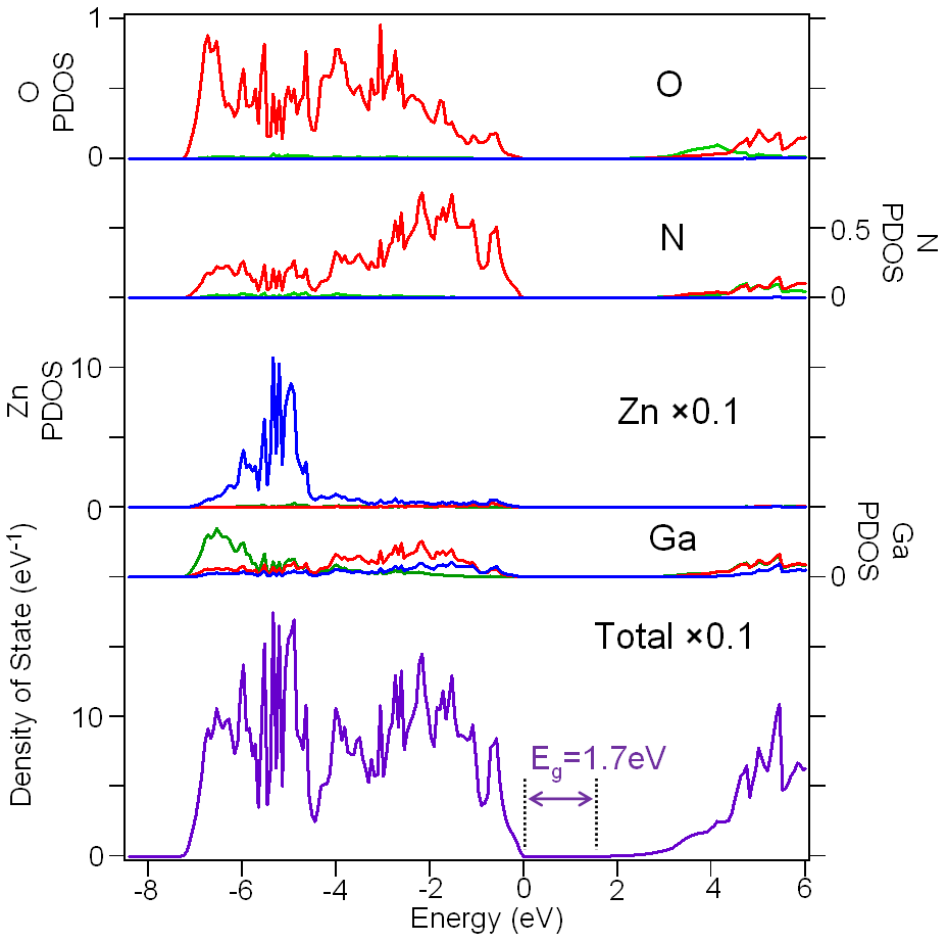

Figure S7. Total density of states (purple curve) and partial density of states (PDOS) of Ga, Zn, O and N atoms in $(Ga_7Zn)(N_7O)$ without split anion sites. Green, red and blue lines denote the s, p and d orbitals, respectively. These curves were obtained by the DFT calculations. The band gap 1.70 eV was estimated from the dispersion curve depicted in Fig. S8B.

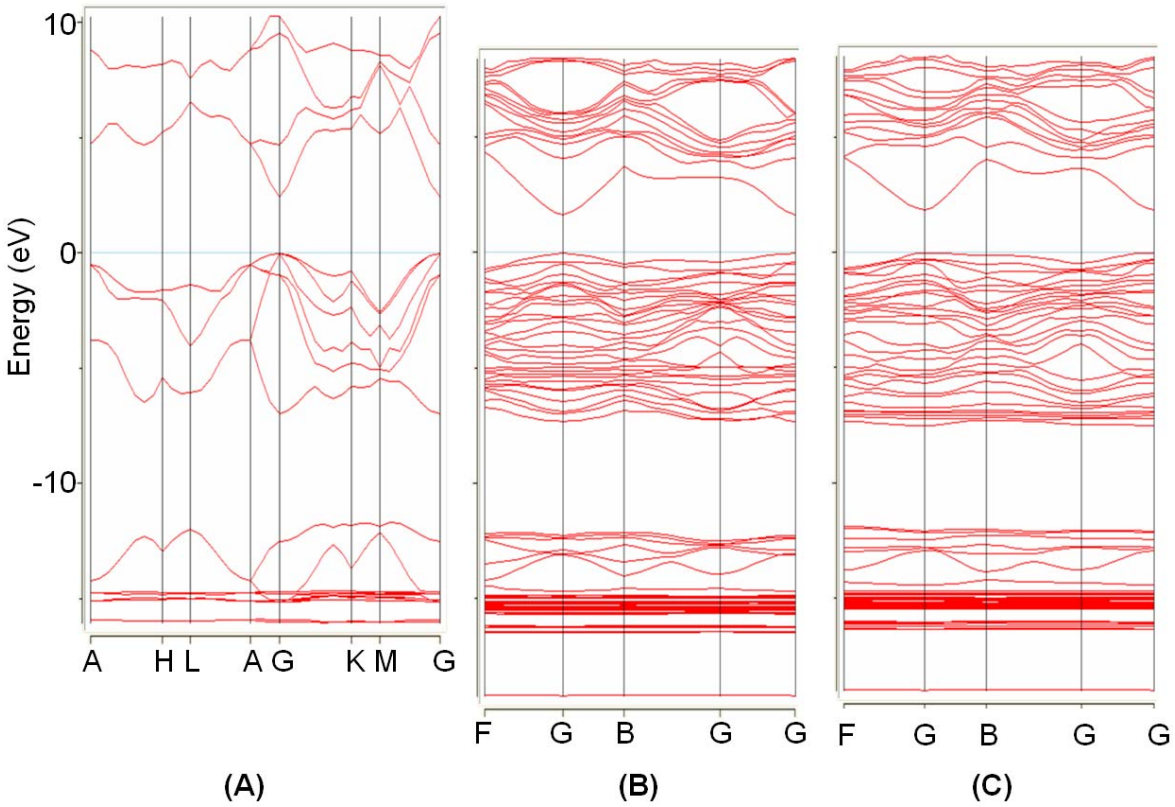

Figure S8. Band structures of GaN (A), $(Ga_7Zn)(N_7O)$ (B) without and (C) with split anion sites. These curves were obtained by the DFT calculations.