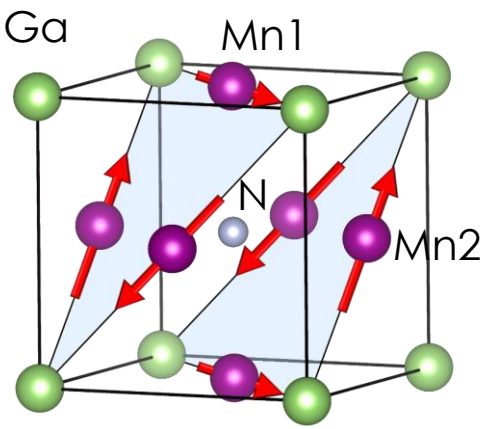
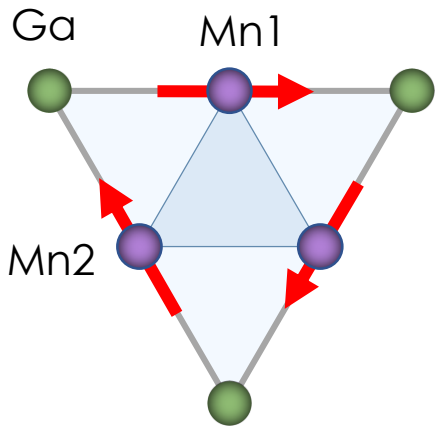
Modeling of the Mn_3GaN/ABO_3 (001) Interface by density functional theory calculations

Bulk Mn_3GaN: Ground state





Computational methods

- VASP+PAW+GGA
- E_{cutoff}: 550 eV
- Kpoints: 16×16×16

	a (Å)	m (μ_B/Mn)
This work	3.866	2.438
Experiment	3.886 [1]	2.68(5) [1]
Previous theory	3.86 [2,3]	2.4 [2], 2.43 [3]

Calculated Mn magnetic moments (μ_B)

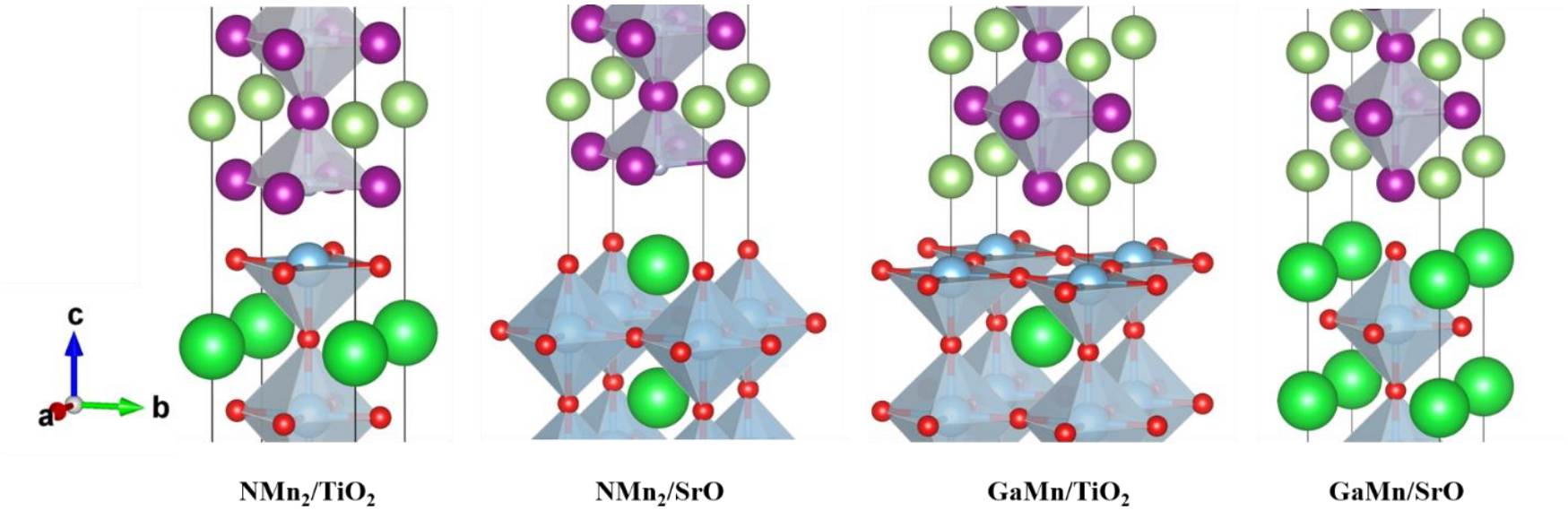
	m_x	m_y	m_z	m_{Tot}
Mn1	1.724	1.724	0	2.438
Mn2	-1.724	0	1.724	2.438
	0	-1.724	-1.724	2.438

[1] K. Shi et al., Adv. Mater. 28, 3761 (2016)
[2] P. Lukashev et al., PRB 78, 184414 (2008)
[3] J. Zemen et al., arXiv:1512.03470 (2015)

Ground state of bulk $GaNMn_3$ successfully reproduced

$Mn_3GaN/SrTiO_3$: Interface stability

$4GaNMn_3{\cdot}NMn_2/4SrTiO_3{\cdot}TiO_2$ $4GaNMn_3{\cdot}NMn_2/4SrTiO_3{\cdot}SrO$ $4GaNMn_3{\cdot}GaMn/4SrTiO_3{\cdot}TiO_2$ $4GaNMn_3{\cdot}GaMn/4SrTiO_3{\cdot}SrO$



NMn_2/TiO_2 NMn_2/SrO $GaMn/TiO_2$ $GaMn/SrO$

- VASP+PAW+GGA
- E_{cutoff}: 550 eV
- Kpoints: 16×16×1
- $a_{inplane}=a_{SrTiO3}$=3.945 Å
- Γ_{5g} non-collinear magnetism

$$\Delta E_{Mn_2N\_TiO_2} = (E_{supercell} - 4E_{Mn_3GaN} - 4E_{SrTiO_3} - E_{Ti} - 2E_O - E_N - 2E_{Mn})/2,$$

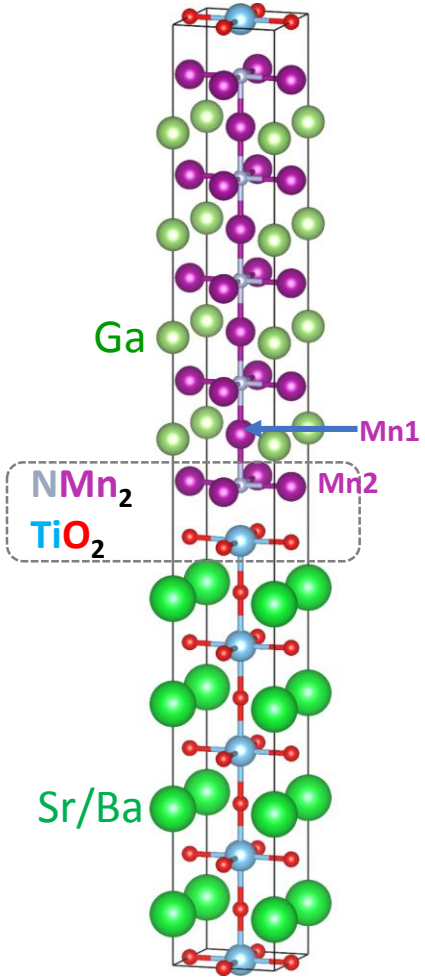
$$\Delta E_{Mn_2N\_SrO} = (E_{supercell} - 4E_{Mn_3GaN} - 4E_{SrTiO_3} - E_{Sr} - E_O - E_N - 2E_{Mn})/2,$$

$$\Delta E_{GaMn\_TiO_2} = (E_{supercell} - 4E_{Mn_3GaN} - 4E_{SrTiO_3} - E_{Ti} - 2E_O - E_{Ga} - E_{Mn})/2,$$

$$\Delta E_{GaMn\_SrO} = (E_{supercell} - 4E_{Mn_3GaN} - 4E_{SrTiO_3} - E_{Sr} - E_O - E_{Ga} - E_{Mn})/2$$

Interface structure	NMn_2/TiO_2	NMn_2/SrO	$GaMn/TiO_2$	$GaMn/SrO$
ΔE (eV/interface)	−4.785	−2.295	−2.965	−2.187

$Mn_3GaN/ATiO_3$ (A=Sr,Ba): Interface structure



$\Delta E = -0.3$ eV/Å^2
Stable

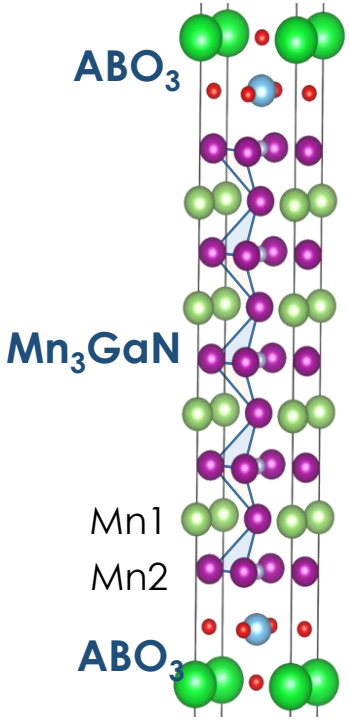
Interfacial Mn2-O and Mn2-Ti distances along the *z* direction for $GaNMn_3/ATiO_3$ (A=Sr, Ba). *P=0* corresponds to $GaNMn_3/SrTiO_3$, while *P>0* and *P<0* correspond to $GaNMn_3/BaTiO_3$ with polarization pointing toward to and away from the interface, respectively.

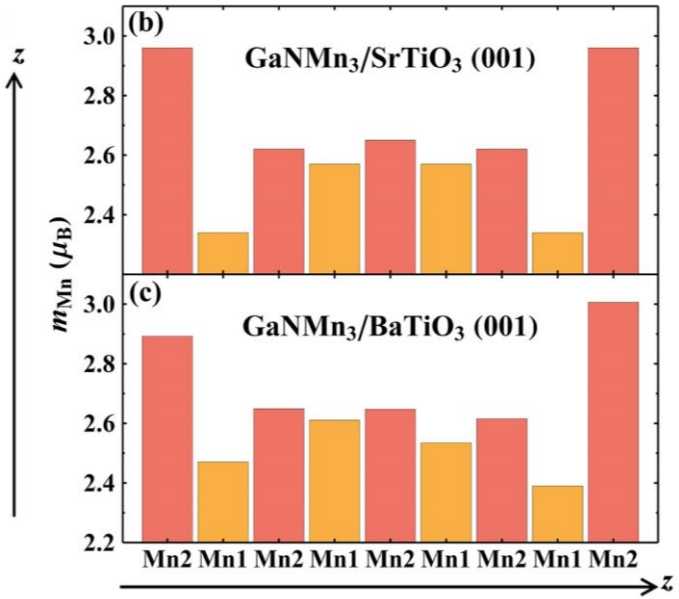
	Mn2-O z-distance (Å)	Mn2-Ti z-distance (Å)	Ti-N z-distance (Å)
$GaNMn_3/SrTiO_3 (P = 0)$	2.087	2.138	2.096
$GaNMn_3/BaTiO_3 (P > 0)$	2.074	1.983	2.015
$GaNMn_3/BaTiO_3 (P < 0)$	2.065	2.253	2.173

$Mn_3GaN/ATiO_3$ (A=Sr,Ba): Magnetsim

The calculated magnetic moments (μ_B) of Mn atoms in $GaNMn_3/ATiO_3$ (A=Sr, Ba).

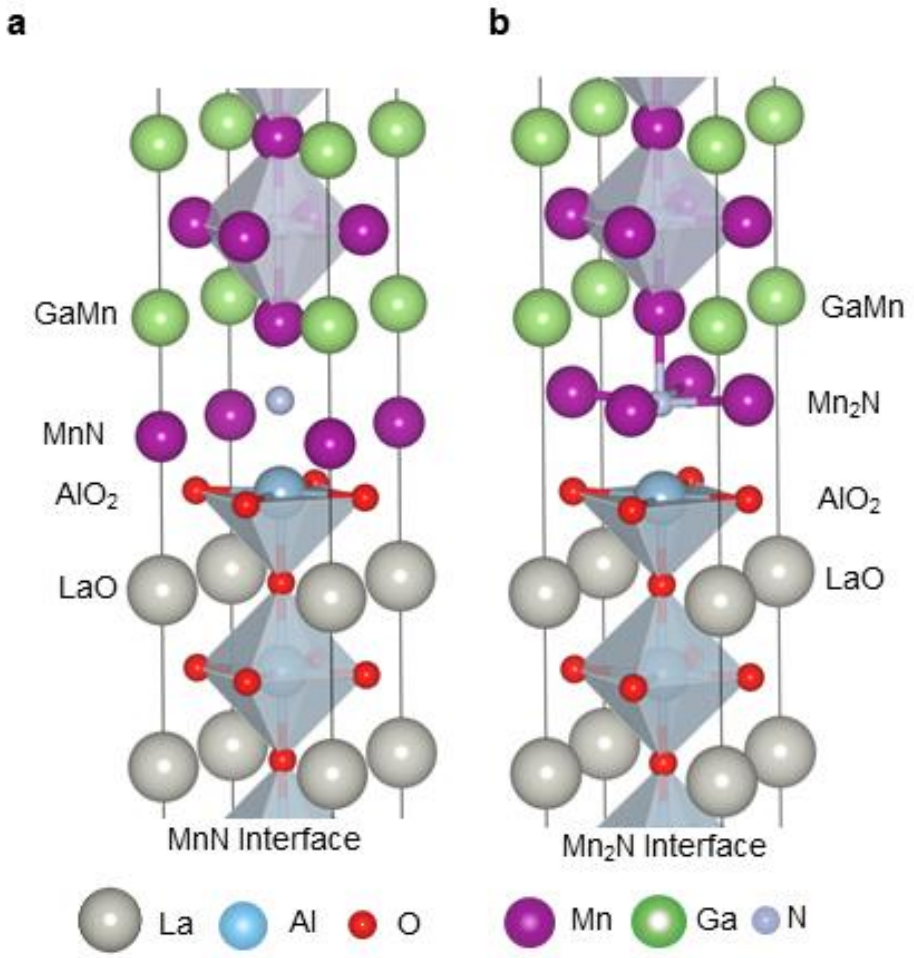
	$GaNMn_3/SrTiO_3$				$GaNMn_3/BaTiO_3$			
	m_x	m_y	m_z	m_{total}	m_x	m_y	m_z	m_{total}
Mn2	0.299	-1.927	-2.227	2.960	-0.732	1.575	2.306	2.887
	-1.929	0.298	2.225	2.960	1.576	-0.732	-2.306	2.887
Mn1	1.659	1.658	0.001	2.345	-1.746	-1.744	0.002	2.468
Mn2	0.183	-1.753	-1.936	2.618	-0.153	1.788	1.940	2.643
	-1.754	0.182	1.935	2.618	1.786	-0.157	-1.942	2.643
Mn1	1.814	1.814	0.001	2.565	-1.839	-1.837	0.002	2.599
Mn2	0.033	-1.854	-1.887	2.646	-0.063	1.832	1.894	2.636
	-1.854	0.033	1.887	2.646	1.830	-0.066	-1.896	2.636
Mn1	1.814	1.814	0.000	2.565	-1.785	-1.784	0.001	2.524
Mn2	0.183	-1.754	-1.936	2.619	-0.181	1.748	1.929	2.609
	-1.753	0.182	1.936	2.618	1.747	-0.184	-1.930	2.610
Mn1	1.659	1.659	0.000	2.346	-1.684	-1.684	0.001	2.382
Mn2	0.300	-1.927	-2.226	2.959	-0.026	2.112	2.137	3.005
	-1.926	0.301	2.227	2.960	2.111	-0.028	-2.138	3.005





$Mn_3GaN/LaAlO_3$: Interface stability

$4GaNMn_3{\cdot}NMn/4SrTiO_3{\cdot}TiO_2$ $4GaNMn_3{\cdot}NMn_2/4SrTiO_3{\cdot}SrO$

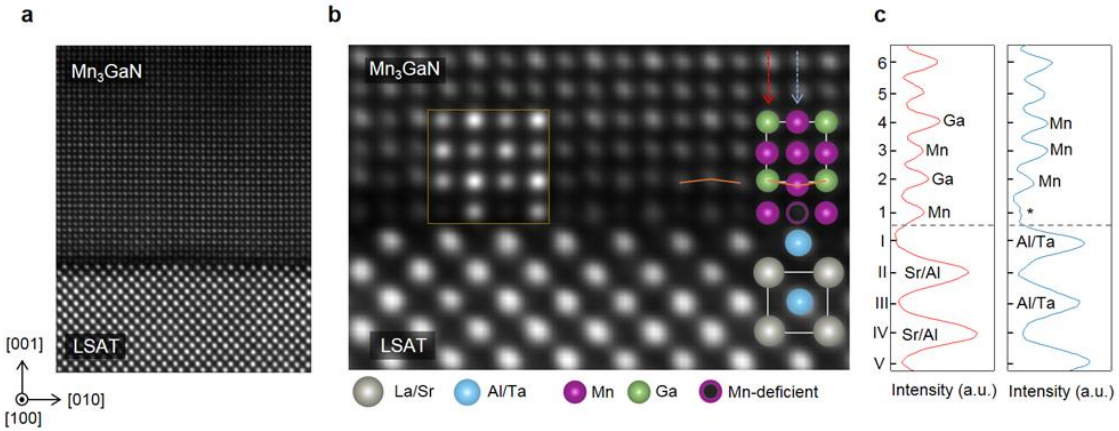


- VASP+PAW+GGA
- E_{cutoff}: 550 eV
- Kpoints: 16×16×1
- $a_{inplane}=a_{Mn3GaN}$=3.867 Å
- Γ_{5g} non-collinear magnetism

$$\Delta E_{MnN\_interface} = (E_{supercell} - 4E_{Mn_3GaN} - 4E_{LaAlO_3} - E_{Al} - E_N - 2E_O)/2,$$
$$\Delta E_{Mn_2N\_interface} = (E_{supercell} - 4E_{Mn_3GaN} - 4E_{LaAlO_3} - E_{Al} - E_N - 2E_O - 2E_{Mn})/2$$

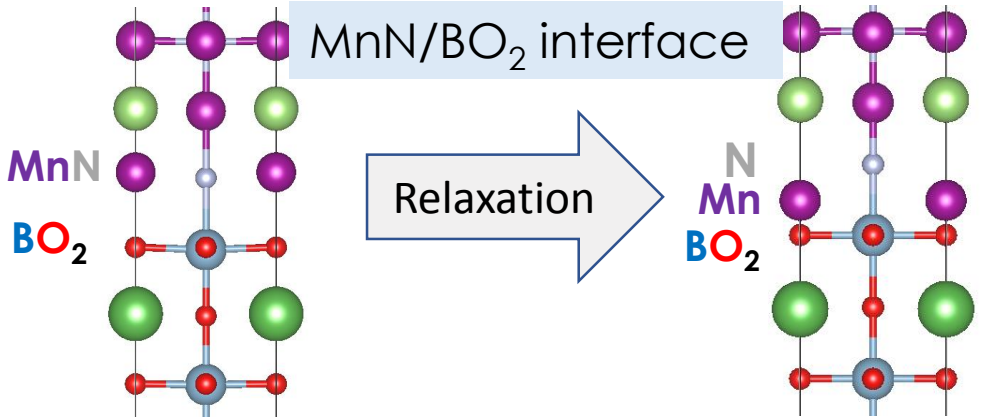
	$Mn_3GaN/LaAlO_3$ (MnN interface)	$Mn_3GaN/LaAlO_3$ (Mn_2N interface)
ΔE (eV/interface)	−0.058	−2.265

Mn_3GaN: Tests of different interfacial configuration



Experiment:

- MnN interfacial monolayer
- GaMn puckered layer

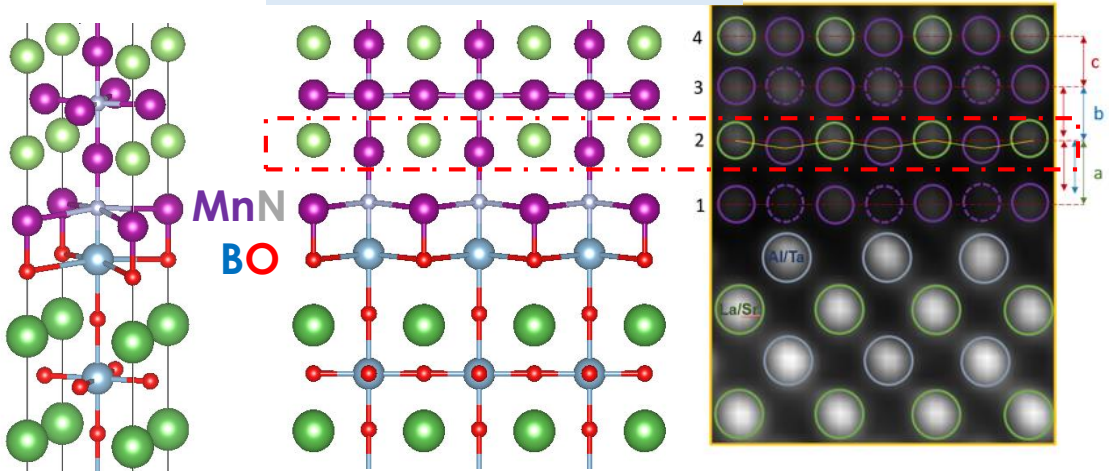


- VASP+PAW+GGA
- E_{cutoff}: 550 eV
- Kpoints: 16×16×1
- Non-magnetism

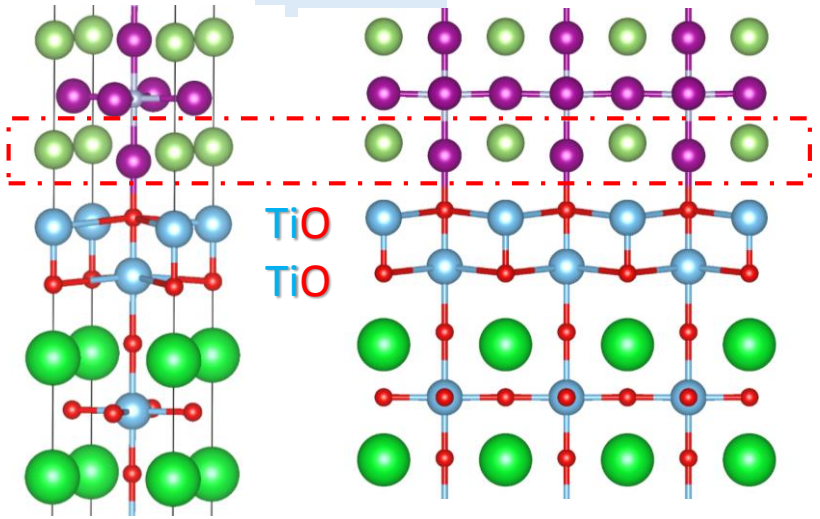
Non-magnetic MnN/BO2 interface doesn't have puckered GaMn layer

Mn_3GaN: Tests of different interfacial configuration

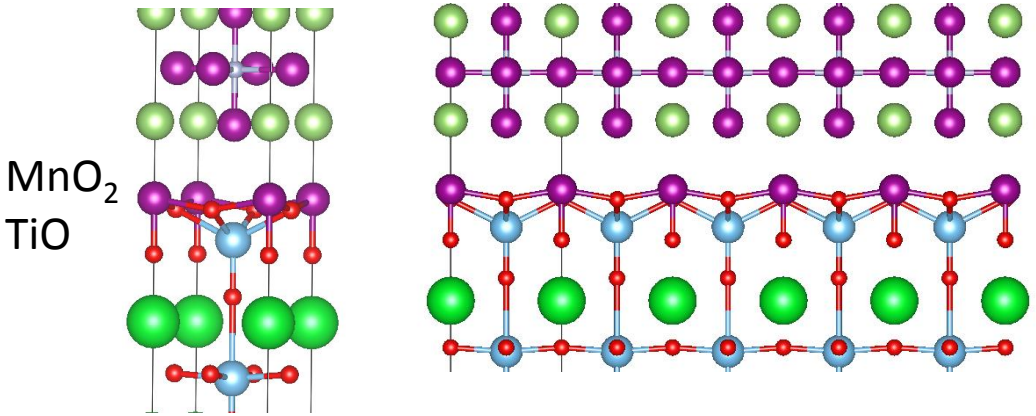
MnN/BO interface



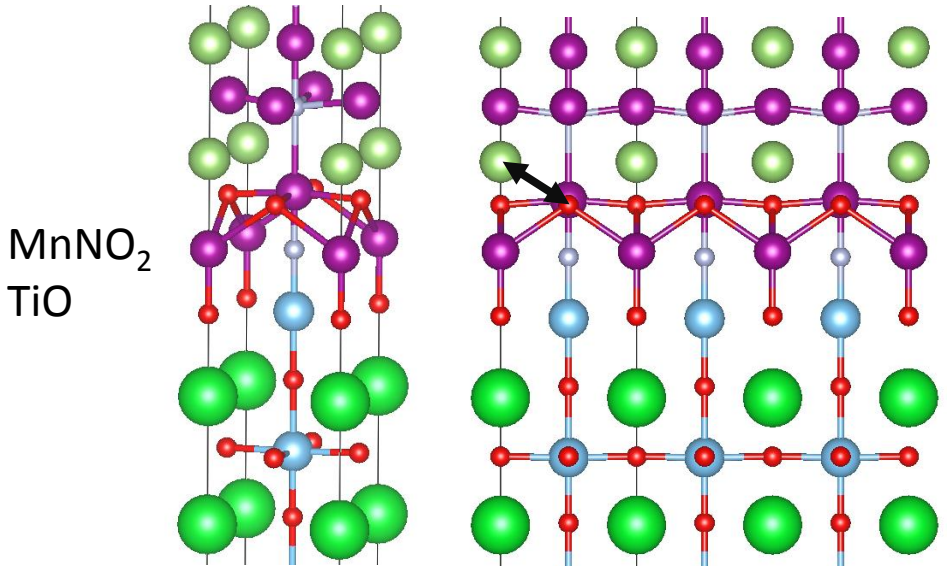
TiO/TiO



MnO_2/TiO interface



$MnNO_2$/TiO



$Mn_3GaN/LaAlO_3$: N rich interfaces

Fix a=3.764 Å (a_{GGA} of non magnetic bulk $GaNMn_3$), relax c without magnetism

Mn_2N monolayer

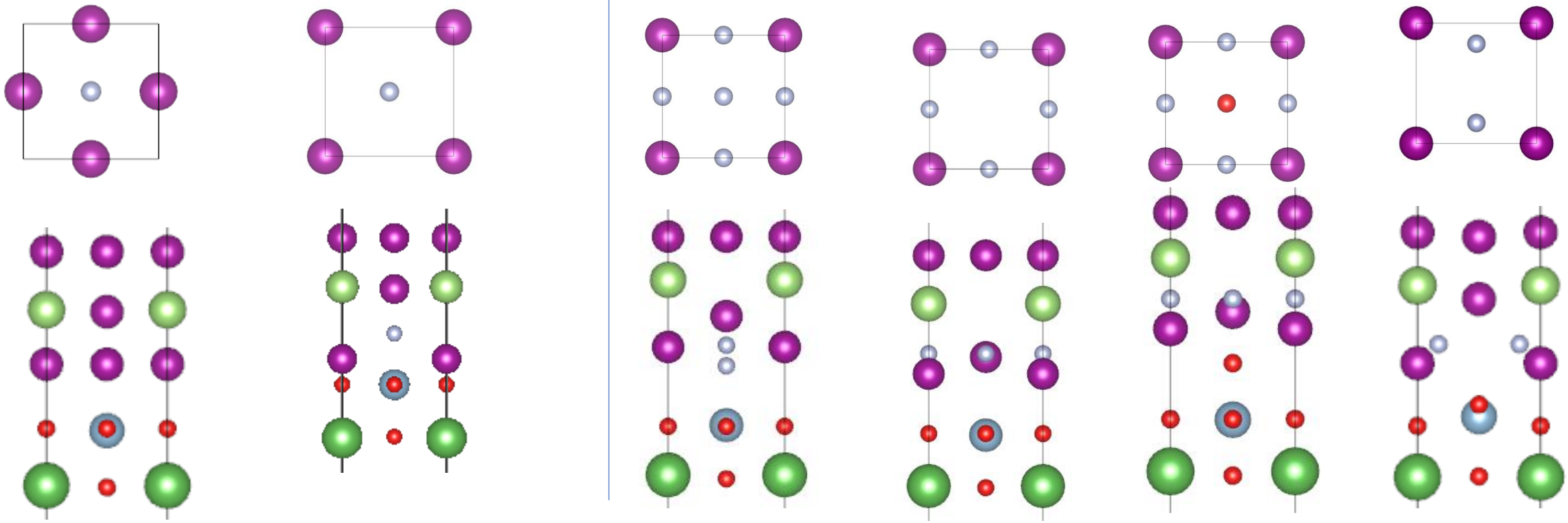
Experimentally proposed MnN monolayer

MnN_3

MnN_2

MnN_2O

MnN_2-2

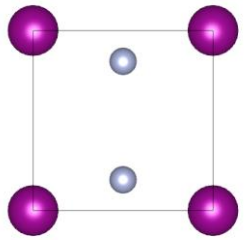


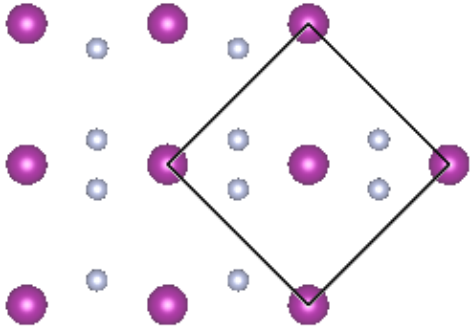
- **MnN_2-2 seems to be a reasonable model to reproduce the puckering**

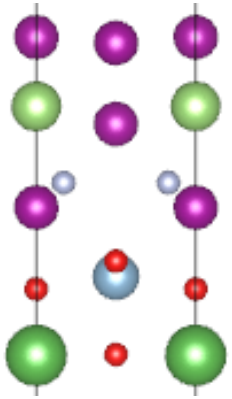
$Mn_3GaN/LaAlO_3$: N rich interfaces

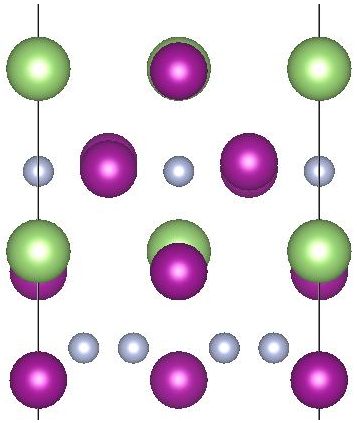
MnN_2-2

$\sqrt{2} \times \sqrt{2}$ cell





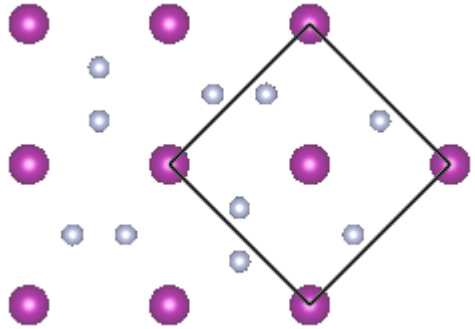


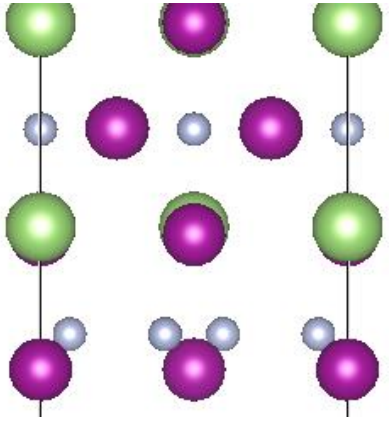


Puckering

MnN_2-3

$\sqrt{2} \times \sqrt{2}$ cell



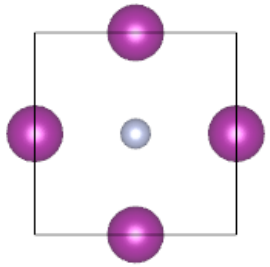


Weaker puckering

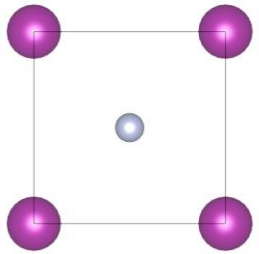
- **MnN_2-3 show weaker puckering structure**

$Mn_3GaN/LaAlO_3$: N rich interfaces

Mn_2N

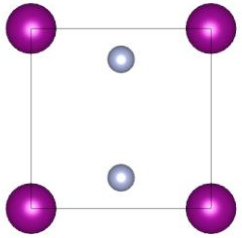


MnN

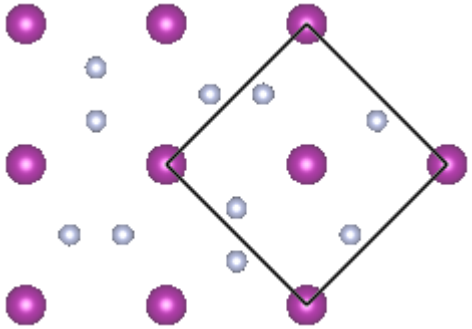


- VASP+PAW+GGA
- E_{cutoff}: 550 eV
- Kpoints: 16×16×1
- Non-magnetism
- VCA for O deficiency

MnN_2-2



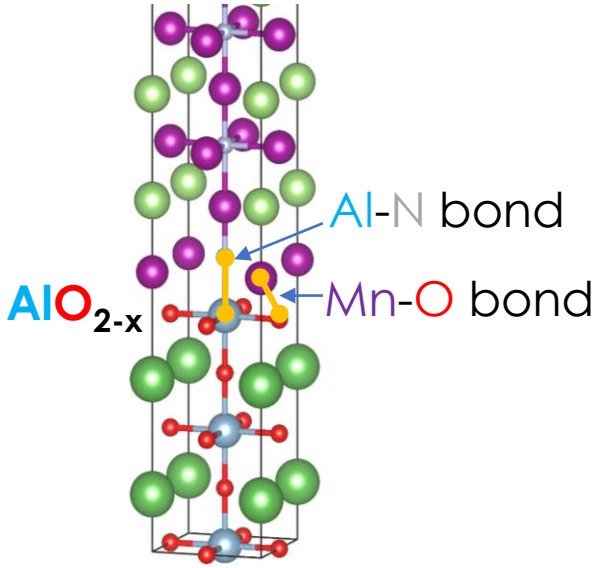
MnN_2-3



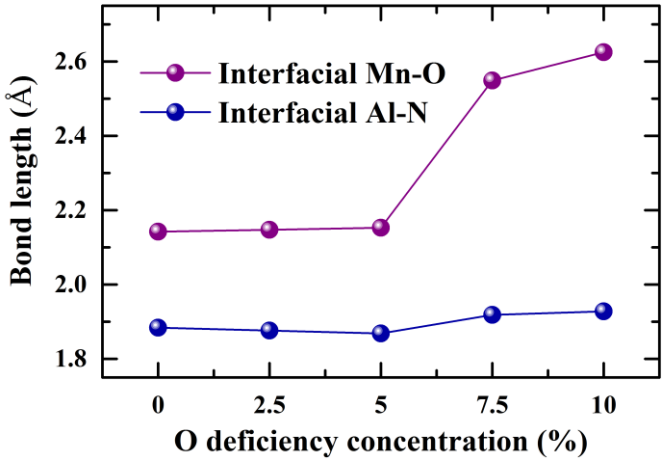
	Formation energy (eV/interface)
Mn2N-supercell (Mn2N/AlO2)	-5.676
MnN-supercell (MnN/AlO2)	-1.459
MnN2-2-supercell	-1.638
MnN2-3-supercell /2	-1.205

Mn_2N structure is more energy favorable

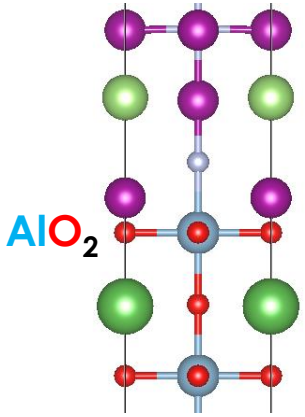
$Mn_3GaN/LaAlO_3$: O deficiency

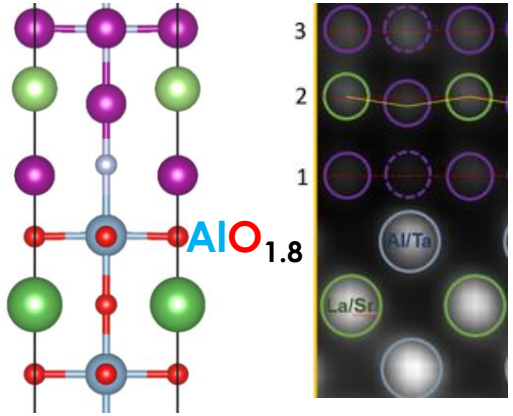


O deficiency simulated by virtual crystal approximation (VCA)



- VASP+PAW+GGA
- E_{cutoff}: 550 eV
- Kpoints: 16×16×1
- Non-magnetism
- VCA for O deficiency

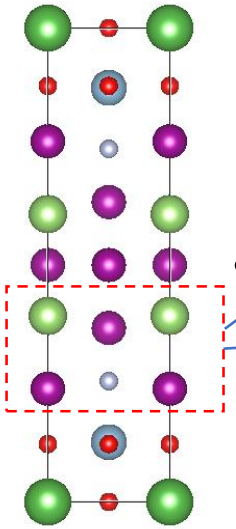




	Mn-O bond length (Å)	Al-N bond length (Å)
MnN/AlO_2	2.142	1.884
$MnN/AlO_{1.95}$	2.147	1.876
$MnN/AlO_{1.9}$	2.153	1.869
$MnN/AlO_{1.85}$	2.549	1.919
$MnN/AlO_{1.8}$	2.625	1.928

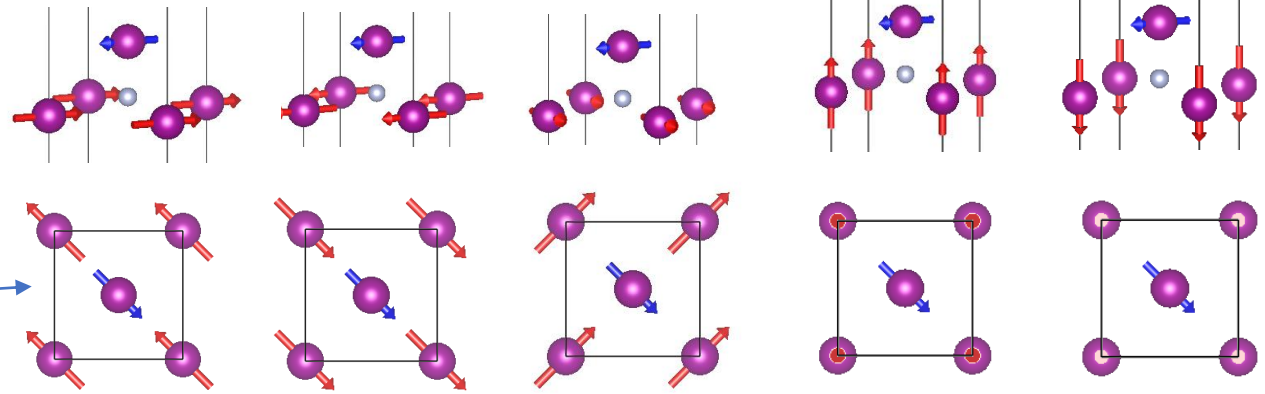
- Interfacial structure reproduced with MnN/AlO_{2-X} configuration

$Mn_3GaN/LaAlO_3$: Magnetsim in small cell



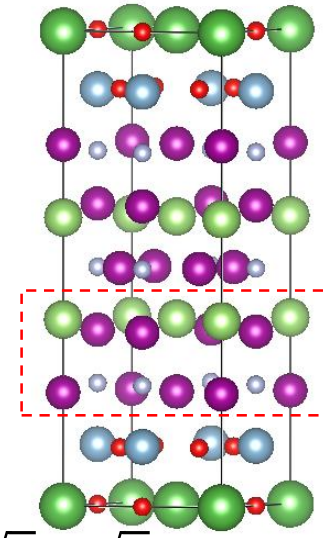
Side view

Top view



E = 0 eV | E = 0.075 eV | E = 0.049 eV | E = 0.068 eV | E = 0.064 eV

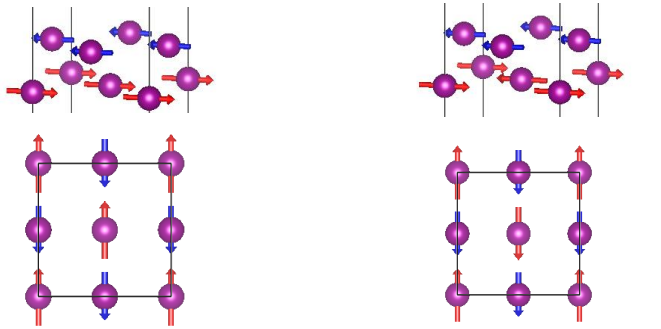
- VASP+PAW+GGA
- E_{cutoff}: 550 eV
- Kpoints: 16×16×1
- $a_{inplane}=a_{Mn3GaN}=3.867$ Å
- Γ_{5g} non-collinear magnetism



$\sqrt{2} \times \sqrt{2} \times 1$ supercell

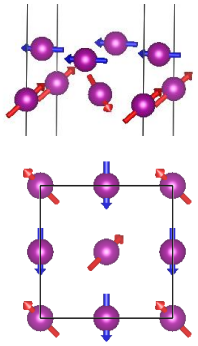
Side view

Top view



E=0 eV

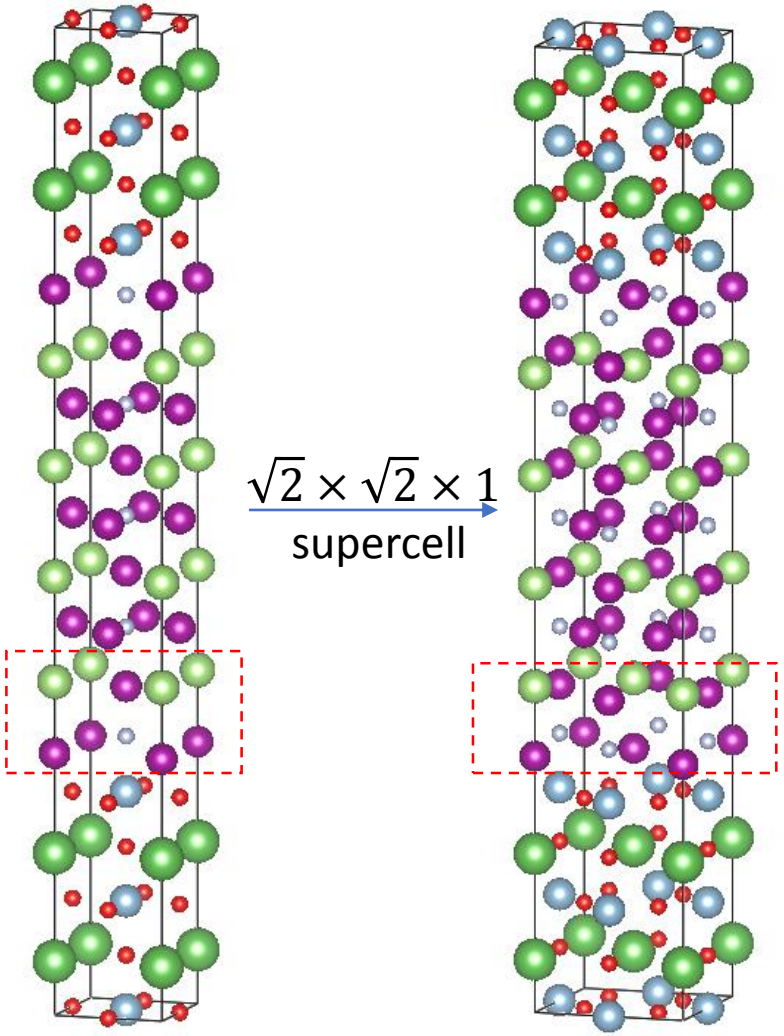
E=0.0033 eV

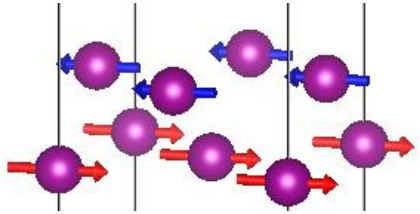


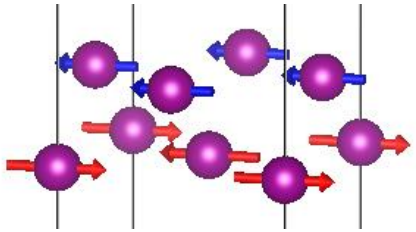
Cannot converge
E fluctuates around 0.05 eV

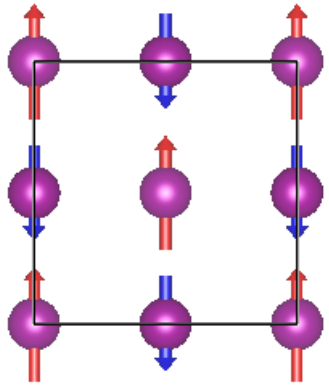
In-plane FM and AFM have the lower energy

$Mn_3GaN/LaAlO_3$: Magnetsim in large cell

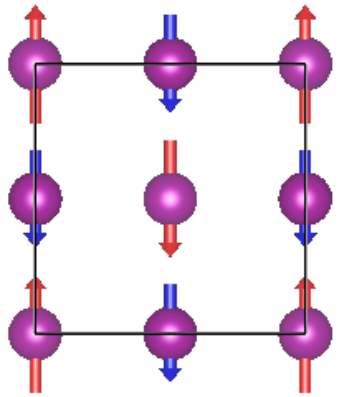








E=0 eV



E=-0.045 eV

The magnetism at interfacial layer should be AFM