Supplemental Material for

Room temperature ferromagnetism properties of $Ga_{14}N_{16-n}Gd_2C_n$ monolayers: A first principle study

Shijian Tian [1], Libo Zhang [3,*], Yuan Liang [1,*], Ruikuan Xie [4], Li Han [3], Shiqi Lan [1], Aijiang Lu [1], Yan Huang [2,*], Huaizhong Xing [1,*], Xiaoshuang Chen [2]

[1]*Department of Optoelectronic Science and Engineering, Donghua University, Shanghai 201620, China*

[2]*State Key Laboratory of Infrared Physics, Shanghai Institute of Technical Physics, Chinese Academy of Sciences, Shanghai 200083, China*

[3]*College of Physics and Optoelectronic Engineering, Hangzhou Institute for Advanced Study, University of Chinese Academy of Sciences, No. 1, Sub-Lane Xiangshan, Xihu District, Hangzhou 310024, China.*

[4]*State Key Laboratory of Structural Chemistry, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences (CAS), Fuzhou, 350002 Fujian, China.*

**Corresponding author*

Email: zhanglibo@ucas.ac.cn (L. B. Zhang)

Email: xinghz@dhu.edu.cn (H. Z. Xing).

Email: yliang@dhu.edu.cn(Y. Liang).

Email: yhuang@mail.sitp.ac.cn (Y. Huang).

To observe the effect of size, the band structures and PDOS of supercells 4×4×1 ($Ga_{14}N_{16}Gd_2$), 5×5×1($Ga_{23}N_{25}Gd_2$) and 6×6×1 ($Ga_{34}N_{36}Gd_2$) are shown at Figure S1. It is found that the band structures are similar, the band gap values for the spin-up channels for the three sizes are 1.99 eV, 2.00 eV and 2.01 eV, respectively, while the spin-down channels are 2.05 eV, 2.07eV, and 2.04 eV, respectively. For the three sizes of $Ga_{14}N_{16}Gd_2$ monolayers, the band structures of spin-up and spin-down channel are similar, with an almost equivalent band gap. The size effect has ignorable influence on the electronic and magnetic properties of the Gd-pair doped GaN monolayer.

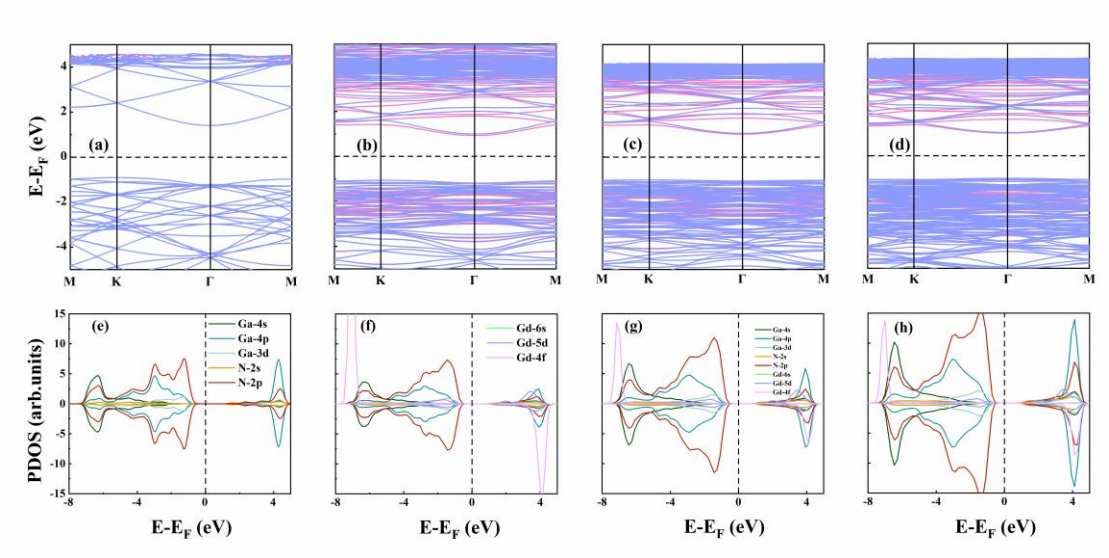

Figure S1. Band structures of (a) $Ga_{16}N_{16}$ (b) $Ga_{14}N_{16}Gd_2$, (c) $Ga_{23}N_{25}Gd_2$ and (d) $Ga_{34}N_{36}Gd_2$ monolayers. The blue and pink solid lines in the band structure represent spin up and spin down channels, respectively. PDOS of (e) $Ga_{16}N_{16}$ (f) $Ga_{14}N_{16}Gd_2$, (g) $Ga_{23}N_{25}Gd_2$ and (h) $Ga_{34}N_{36}Gd_2$ monolayers. The dash lines present the fermi-level which is taken to be 0.

The magnetic coupling between Gd and C atoms are studied by mapping the total energy to the classical Heisenberg model. The nearest-neighboring exchange coupling is considered to associated with the magnetic phase and label it as J. The Hamiltonian can be defined as:

$$H=-J_1 \sum_{<i,j>} S_i S_j \qquad 1$$

In the formula, S is the spin vector; <i, j> is summation over the nearest-neighboring Gd-pair. The energies of magnetic states can be described as:

$$E_{FM}=E_0-3JS^2,\ E_{AFM}=E_0+JS^2$$

E_0 is the reference energy. Thus, the magnetic coupling parameter (J) can be defined as:

$$J=(E_{AFM}-E_{FM})/4S^2 \quad 2$$

The corresponding atomic structure is shown at Figure 1, the positions of the C-atom doping are marked as the numbers inserted in Figure 1a. The E_b, $\triangle E$, M_{total}, J [39] and T_C are studied by PBE method are shown at Table S1 and S2. It can be observed that the E_b and M_{total} of the FM phase are equivalent for a fixed C concentration, indicating that the substituted sites substitution has no impact on the structural stability and magnetic moment. However, the structural stability decreases with the increase of C concentration. For example, when the C atom concentration reaches 12.5% (4 C atoms), the magnetic state is no more stable and the structural stability decreases by 16.49%. It can be seen that the substitutional impurity of C atom has significant effect on the ground state of GaN: Gd monolayers, not all doping scheme maintain positive $\triangle E$ and J. The positive $\triangle E$ and J values indicate the FM phase, which is detected in the following configurations: [M], [1, 3], [M, 6], [M, 8], [2, 3, 5], [M, 5, 7] and [M, 5, 7, 8]. Negative values indicating AFM phases which are detected the following configurations: [1], [4], [5], [8], [1, 2], [5, 7], [M, 1], [M, 2], [1, 2, 3], [5, 6, 7], [1, M, 2], [1, M, 3], [2, M, 5] and [M, 4, 6, 7].

Table S1. The E_b of FM phase, total magnetic moments (M_{total}), ΔE (E_{AFM}-E_{FM}), nearest-neighboring exchange (J), estimated T_C^{FMA} and T_C of $Ga_{14}N_{16}Gd_2$, $Ga_{14}N_{15}Gd_2C_1$, $Ga_{14}N_{14}Gd_2C_2$, $Ga_{14}N_{13}Gd_2C_3$, $Ga_{14}N_{12}Gd_2C_4$, respectively.

	E_b (eV)	M_{total} (μ_B)	△E (meV)	J (meV)	T_C^{FMA} (K)	T_C (K)
$Ga_{14}N_{16}Gd_2$	-5.64	14.00	-4.08	-0.08	-	-
[1]	-5.56	15.00	-3.82	-0.08	-	-
[4]	-5.57	15.00	-10.26	-0.21	-	-
[5]	-5.57	15.00	-4.04	-0.08	-	-
[8]	-5.57	15.00	-1.90	-0.04	-	-
[M]	-5.54	15.00	15.05	0.31	58.22	35.51
[1, 2]	-5.48	16.00	-0.77	-0.02	-	-
[5, 7]	-5.50	16.00	-0.90	-0.02	-	-
[M, 1]	-5.47	16.00	-1767.19	-36.07	-	-
[M, 2]	-5.47	16.00	-1770.46	-36.13	-	-
[1, 3]	-5.48	16.00	14.77	0.30	57.13	34.85
[M, 6]	-5.48	16.00	14.14	0.29	54.70	33.36
[M, 8]	-5.48	16.00	111.81	2.26	428.63	261.46
[2, M, 5]	-5.08	17.00	-2222.17	-45.36	-	-
[1, M, 2]	-5.07	17.00	-2335.62	-47.67	-	-
[1, M, 3]	-5.08	17.00	-2409.58	-49.18	-	-
[1, 2, 3]	-5.09	17.00	-0.49	-0.01	-	-
[5, 6, 7]	-5.12	17.00	-61.21	-1.25	-	-
[2, 3, 5]	-5.10	17.00	51.26	1.05	198.28	120.95
[M, 5, 7]	-5.10	17.00	82.55	1.68	319.32	194.78
[M, 4, 6, 7]	-4.71	18.00	-3.34	-0.07	-	-
[M, 5, 7, 8]	-4.71	18.00	155.12	3.17	600.03	366.02

The PDOS of following examples are shown in Fig. S2, which are used to investigate the underlying mechanisms of the above phenomena: [1], [4], [M, 2], [5, 7], [2, M, 5] and [1, M, 2]. It is shown that one of the factors causing the negative △E and

J is due to the apparently weaker p-d coupling between the Gd-5d and C-2p states near the Fermi level ([1], [4] and [5, 7] in Figure S3). In addition, the energy of the AFM state of the special substitution site is reduced dramatically, resulting in AFM ground state, such as [M, 2], [2, M, 5], [1, M, 2] (shown in Figure S4). In summary, we define the ideal scheme with a stable FM phase and relatively high T_C: one C atoms at M positions and other C atoms bonded without Gd atoms: (M), (M, 8) and (M, 5, 7).

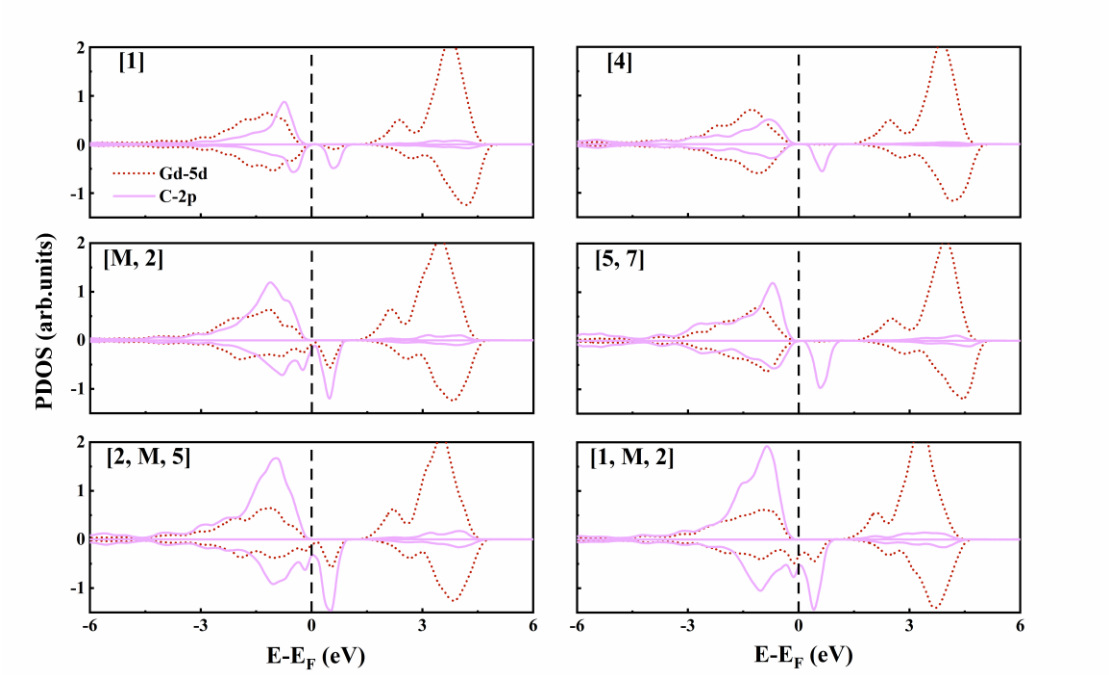

Figure S2. PDOS with FM states of different substitution site: [1], [4], [M, 2], [5, 7], [2, M, 5] and [1, M, 2].

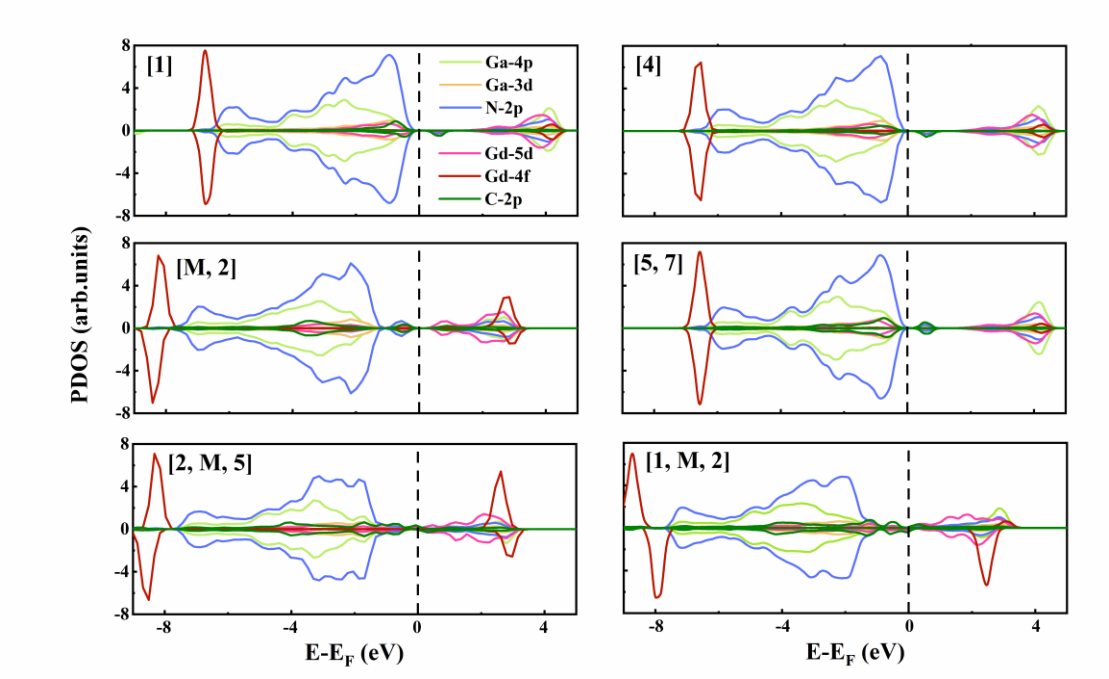

Figure S3. PDOS with AFM states of different substitution site: [1], [4], [M, 2], [5, 7], [2, M, 5] and [1, M, 2].

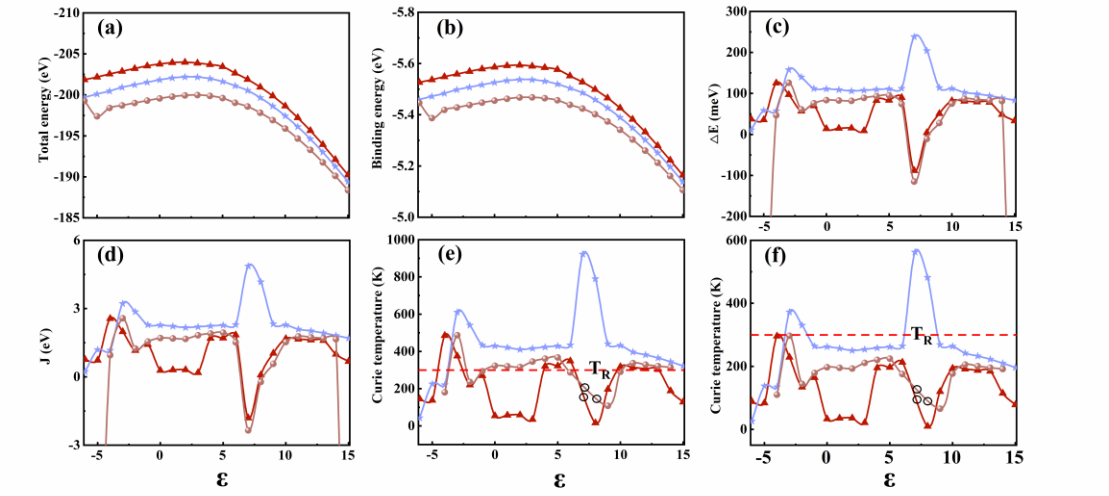

Figure S4. (a) E_{total} of FM phase, (b) E_b, (c) $\triangle E$ (d) J, (e) T_C^{FMA} and (f) T_C of $Ga_{14}N_{16-n}Gd_2C_n$ monolayers computed by the PBE method with SOC. The dark blue (triangle), light blue (pentagram) and brown (circle) lines in the diagram represent $Ga_{14}N_{15}Gd_2C_1$, $Ga_{14}N_{14}Gd_2C_2$ and $Ga_{14}N_{13}Gd_2C_3$ monolayers, respectively. The black circles in the last two figures indicate this position as an AFM state without T_C.

Table S2. A brief summary of the magnetic properties for doped GaN materials.

Materials	Magnetic type	Magnetic moment	T_C	Ref.
$Ga_{35}N_{36}Mn$	FM	$4.0\mu_B$	-	[5]
$Ga_{35}N_{36}Cu_2$	FM	$4.0\mu_B$	-	[14]
$Ga_{16}N_{15}Fe$	FM	$5.0\mu_B$	-	
$Ga_{16}N_{15}Co$	FM	$4.0\mu_B$	-	[15]
$Ga_{16}N_{15}Ru$	FM	$5.0\mu_B$	-	
$Ga_{16}N_{15}Dy$	FM	$5.1\mu_B$	-	[19]
$Ga_{16}N_{14}GdSm$	FM	$12.01\mu_B$	-	[20]
$CrBr_3$	FM	-	34K	[40]
CrI_3	FM	-	45K	[41]
Fe_3GeTe_2	FM	-	130K	[42]
$CrSe_2$	FM	-	65K	[43]
$CrGeTe_3$	FM	-	30K	[44]