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Supporting Information

Tunable Magnetic Order in Two-dimensional Layered $GdGe_2$

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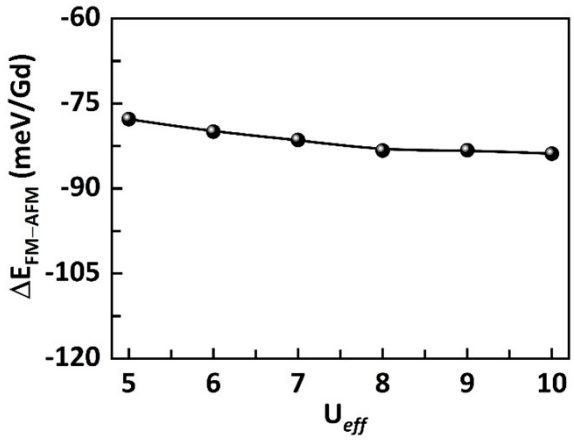

Figure S1. Energy difference between the ferromagnetic (FM) and anti-ferromagnetic (AFM) states of monolayer $GdGe_2$ calculated with different U_{eff}.

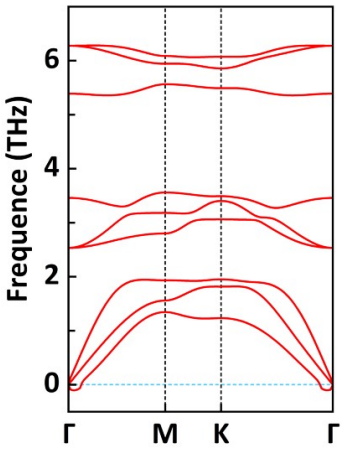

Figure S2. Phonon dispersion of $GdGe_2$ monolayer in the high-symmetry directions of the Brillouin zone.

The function relationships of Young's modulus (Y_{2D}) and Poisson's ratio (ν) with polar angle (φ) are as following:

$$Y_{2D}(\varphi) = \frac{C_{11}C_{22} - C_{12}^2}{C_{11}s^2 + C_{22}c^4 + (\frac{C_{11}C_{22} - C_{12}^2}{C_{66}} - 2C_{12})c^2s^2}$$

$$\nu(\varphi) = \frac{\left(C_{11} + C_{22} - \frac{C_{11}C_{22} - C_{12}^2}{C_{66}}\right)c^2s^2 - C_{12}(s^4 + c^4)}{C_{11}s^2 + C_{22}c^4 + (\frac{C_{11}C_{22} - C_{12}^2}{C_{66}} - 2C_{12})c^2s^2}$$

where φ is the polar angle relative to the x-axis, s and c are $sin\varphi$ and $cos\varphi$, respectively.

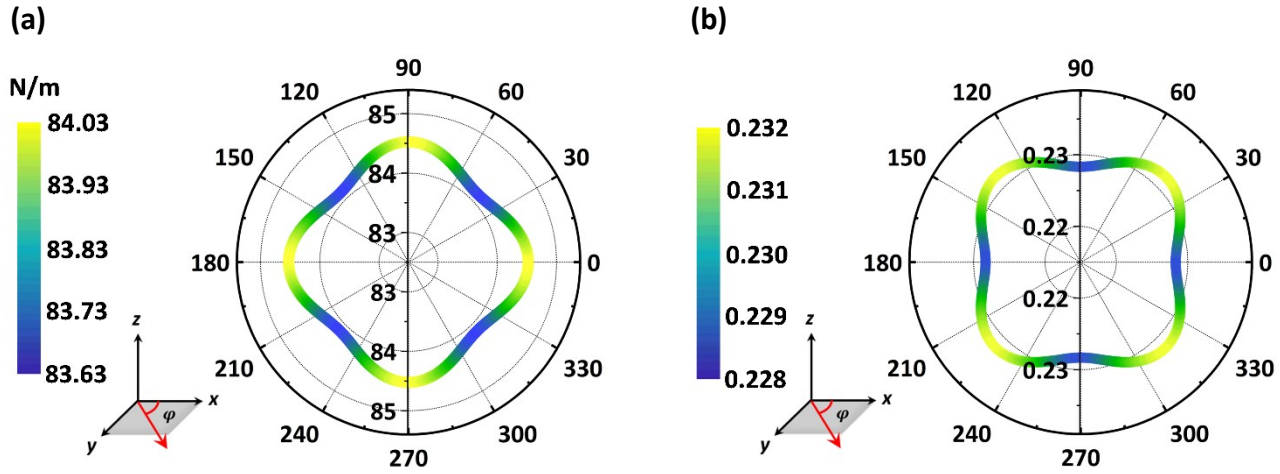

Figure S3. The evolution of (a) Young's modulus and (b) Poisson's ratio with respect to the polar angle (φ) for monolayer $GdGe_2$.

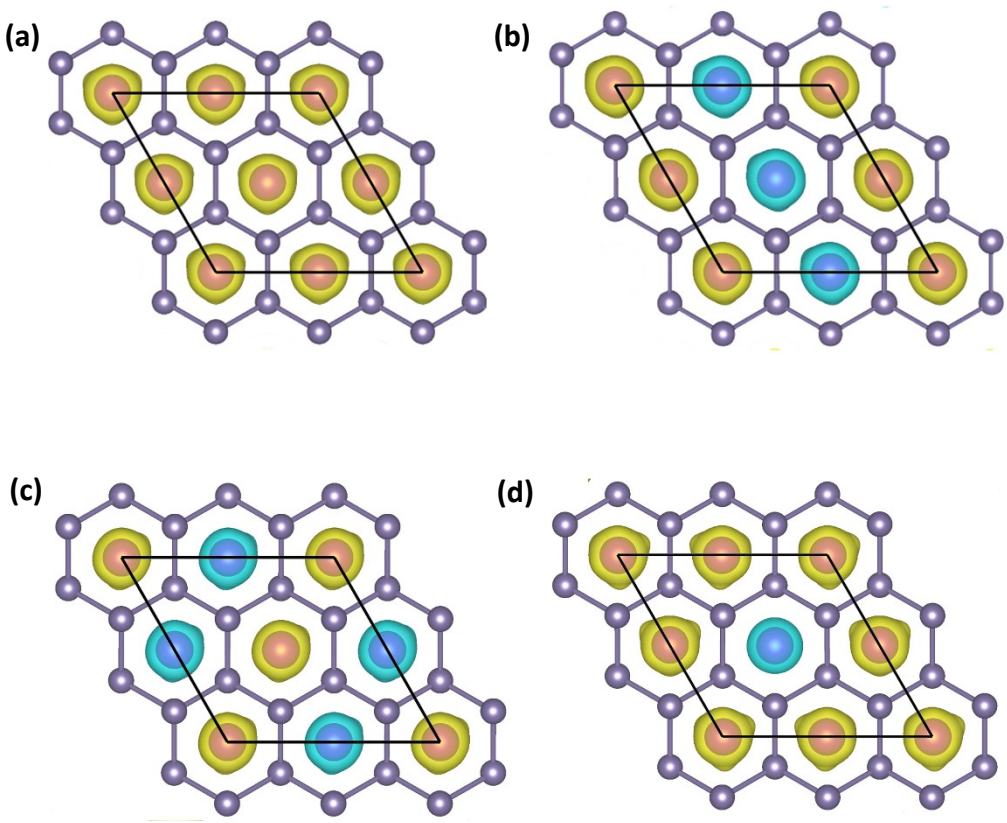

Figure S4. Spin-resolved charge density of $GdGe_2$ monolayer in (a) ferromagnetic (FM), two anti-ferromagnetic (AFM), (b) G-AFM, (c) C-AFM, and (d) ferrimagnetic (FIM) configurations, respectively. The yellow and cyan colors represent the spin-up and spin-down charge, respectively. The isosurface value is set as 0.008 e $Å^{-3}$.

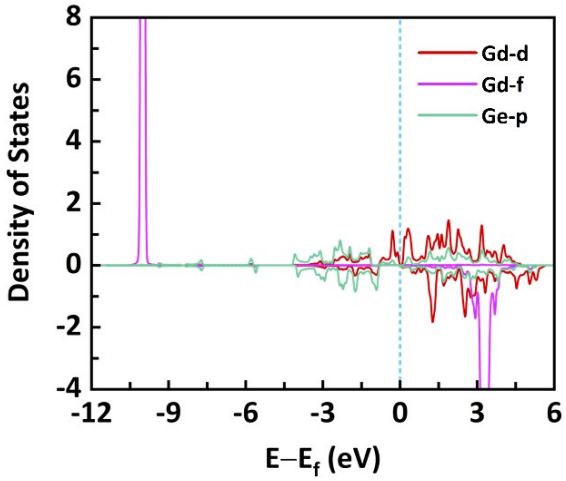

Figure S5. Orbital-resolved projected density of states of Gd and Ge atoms for monolayer $GdGe_2$ calculated by PBE+U method.

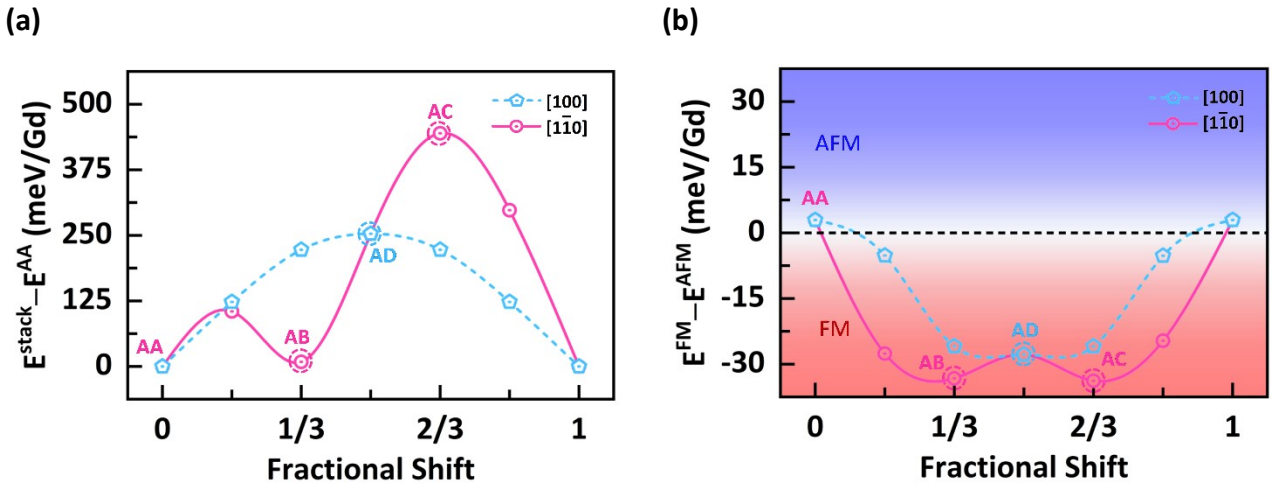

Figure S6. (a) Evolution of the interlayer stacking energy and (b) the interlayer exchange energy of bilayer $GdGe_2$ with respect to the relative displacement of one layer to the other along high-symmetry [100] and [$1\bar{1}0$] directions after stacking-constraint atomic relaxation. Positive (negative) value in (b) represents the anti-ferromagnetic (AFM) (ferromagnetic, FM) interlayer exchange interaction.

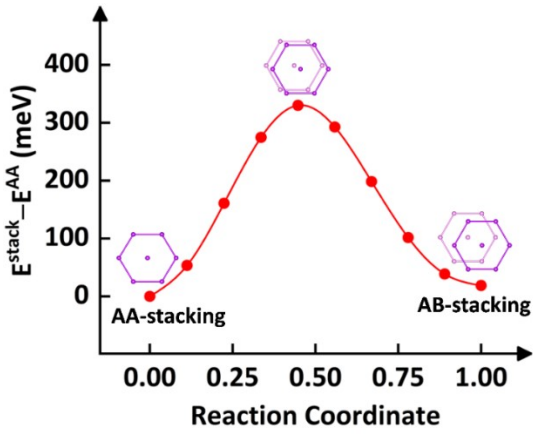

Figure S7. Minimum energy path for the stacking order transition between AA-stacking and AB-stacking bilayer $GdGe_2$ calculated by CI-NEB method.

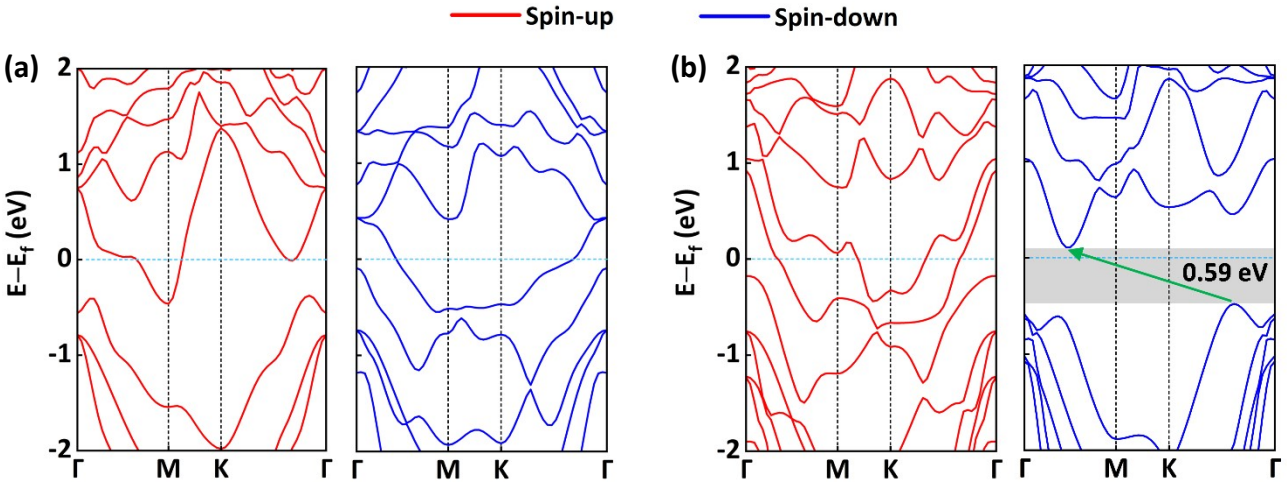

Figure S8. Spin-resolved band structures of (a) AA-stacking and (b) AB-stacking bilayer $GdGe_2$ calculated by HSE06 method.

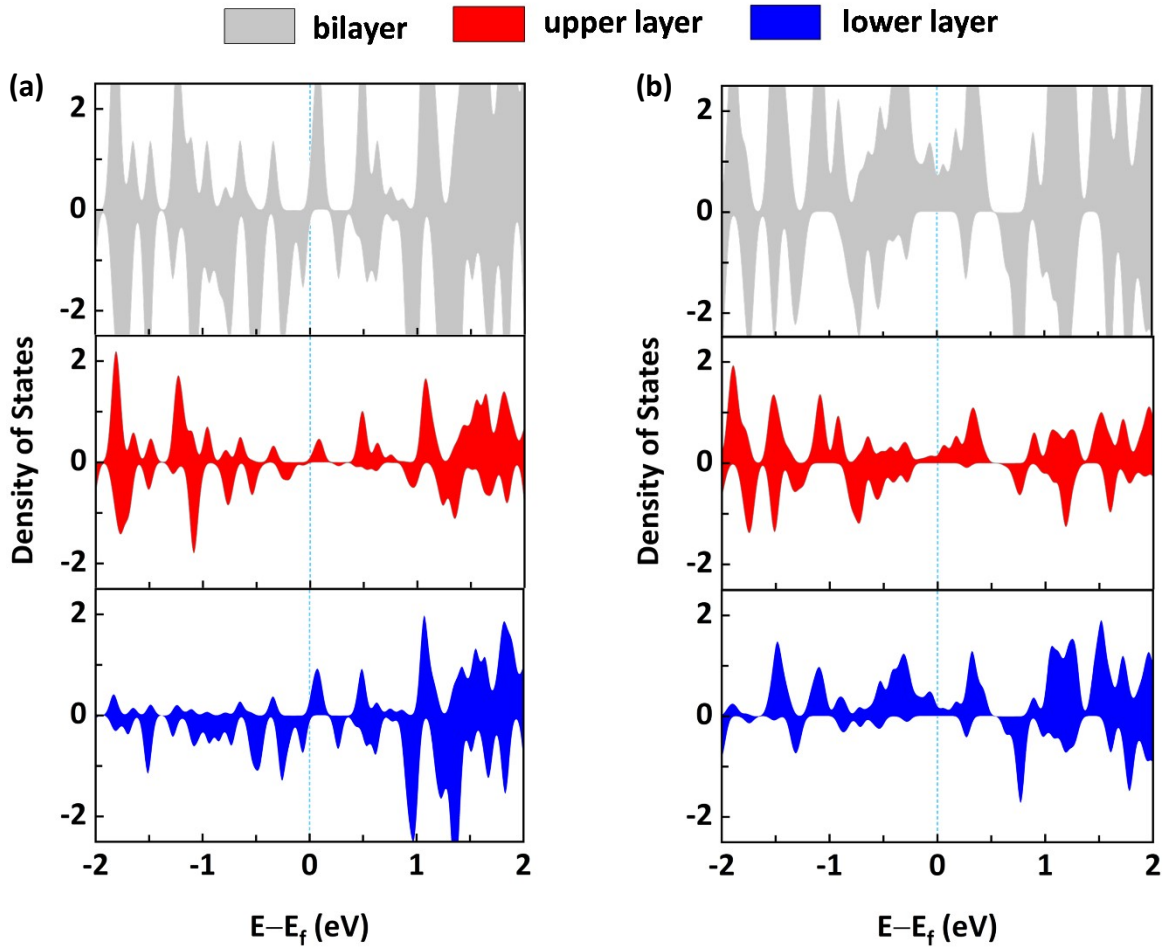

Figure S9. The total density of states and layer-resolved partial density of states for (a) AA- and (b) AB-stacking bilayer $GdGe_2$.

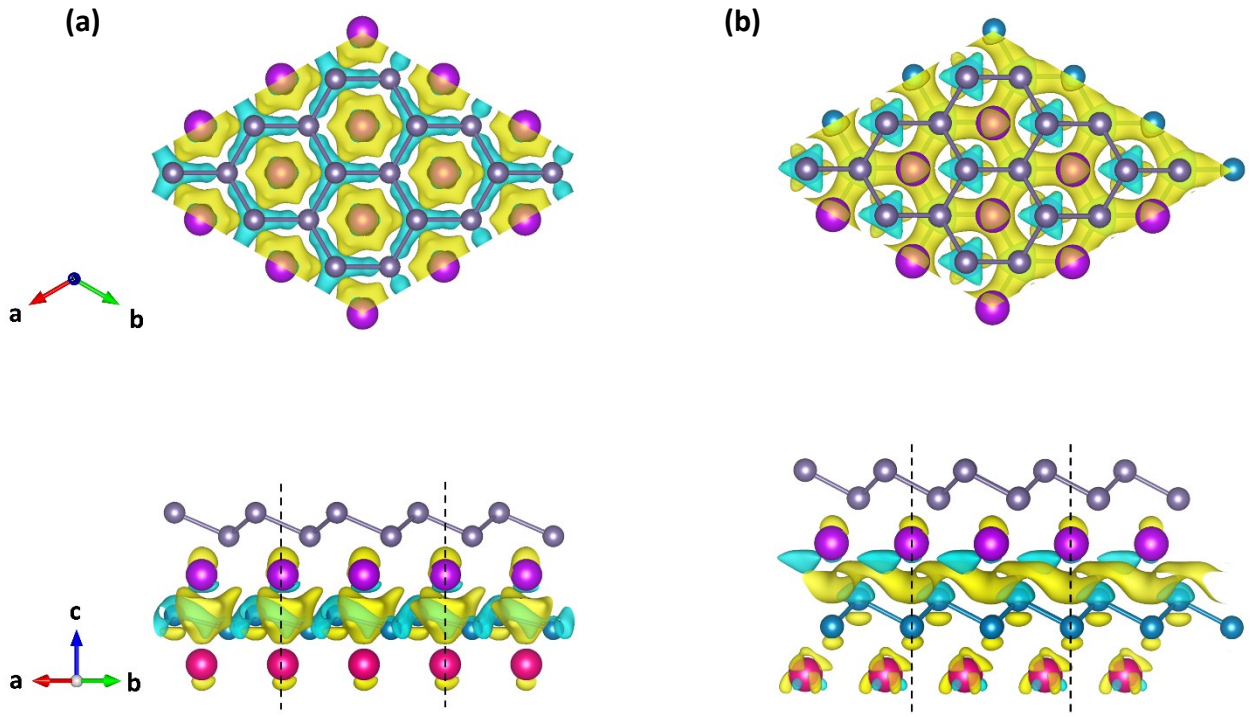

Figure S10. Top and side views of differential charge density for (a) AA-stacking and (b) AB-stacking bilayer $GdGe_2$. The isosurface value is set as 0.002 e $Å^{-3}$. The yellow and cyan colors represented the charge accumulation and depletion, respectively.

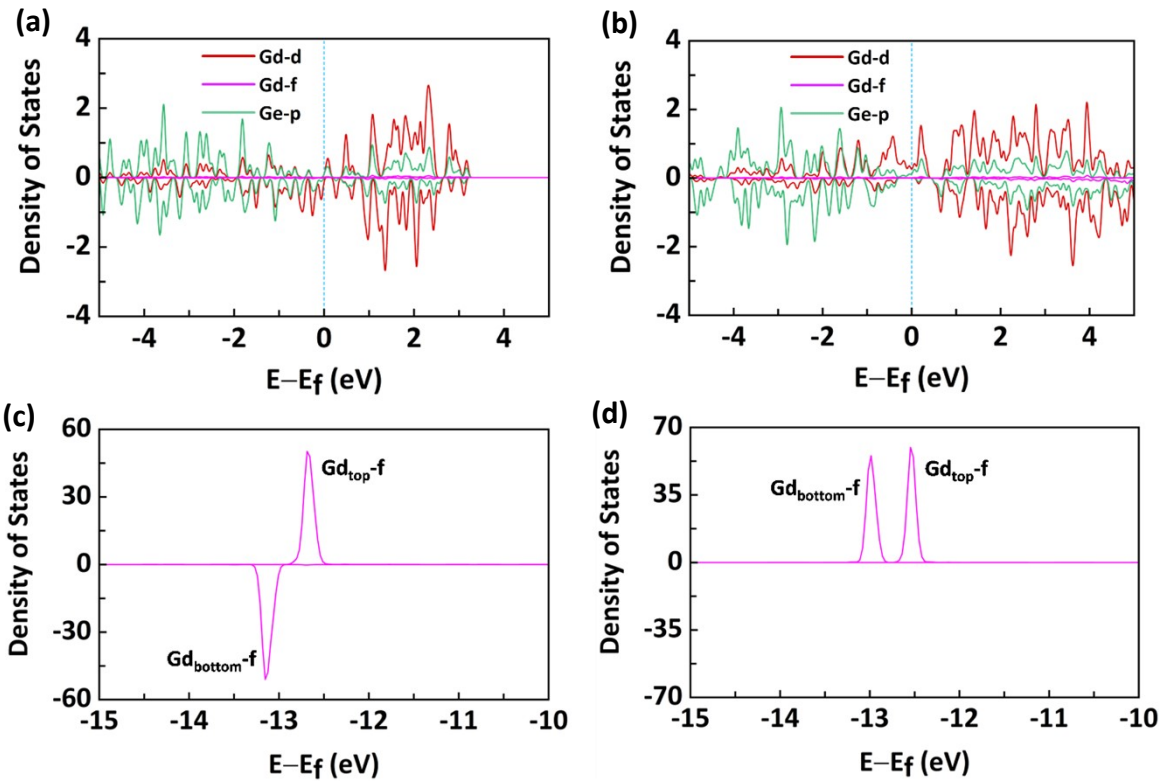

Figure S11. Orbital-resolved projected density of states of Gd and Ge atoms in (a) AA-stacking and (b) AB-stacking bilayer $GdGe_2$ calculated by HSE06 method. The distribution of Gd-f orbitals in (c) AA-stacking and (d) AB-stacking bilayer $GdGe_2$ calculated by HSE06 method. Gd_{top} (Gd_{bottom}) represents the Gd atoms of top (bottom) layer in bilayer $GdGe_2$.

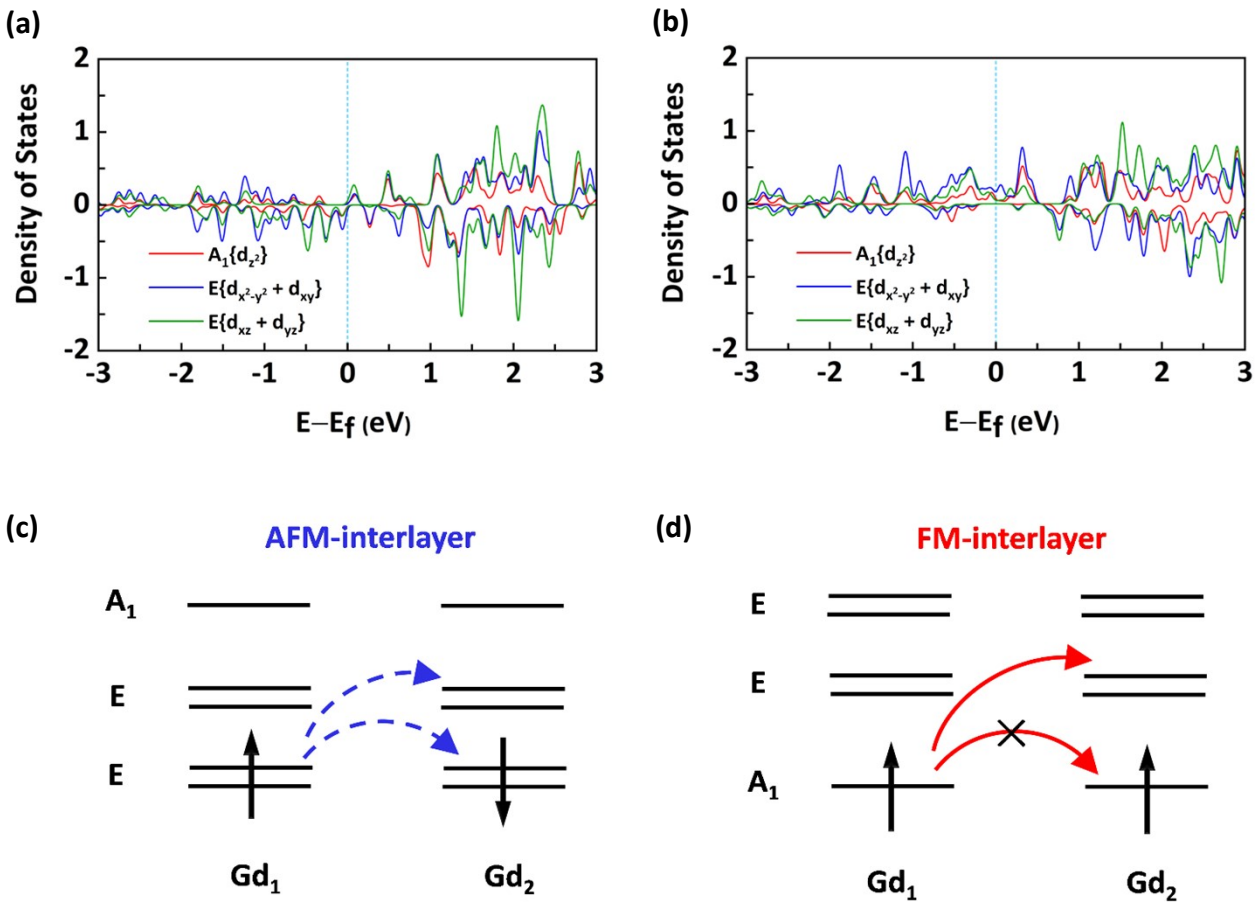

Figure S12. Orbital-resolved projected density of states of Gd atom in (a) AA-stacking and (b) AB-stacking bilayer $GdGe_2$ calculated by HSE06 method, respectively. (c) and (d) Schematic diagram showing the hopping mechanism of Gd-5d electrons for anti-ferromagnetic (AFM) and ferromagnetic (FM) interlayer exchange interactions, respectively.

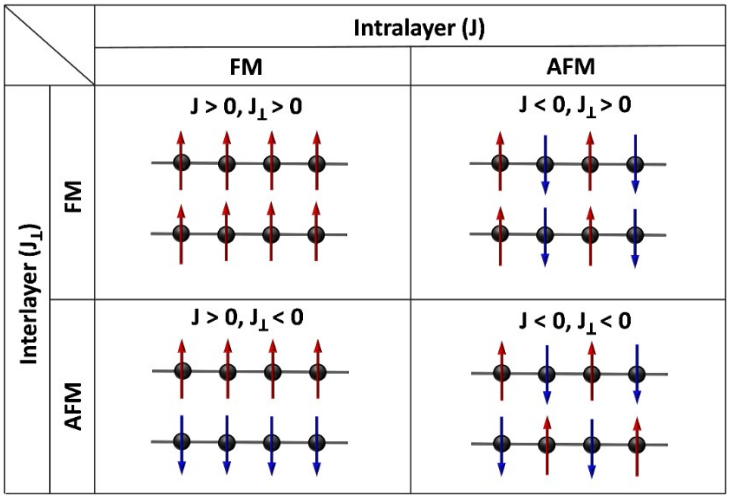

Figure S13. Schematic diagram showing the four magnetic configurations of bilayer $GdGe_2$ with considering the intralayer and interlayer magnetic couplings.

Considering with the interlayer nearest neighboring (NN) and second NN (2NN) magnetic couplings, the spin-Hamiltonian of the bilayer $GdGe_2$ is calculated as:

$$H = -\sum_{nm} J\vec{S}_n \cdot \vec{S}_m - \sum_{ij} J_{1\perp}\vec{S}_i \cdot \vec{S}_j - \sum_{ik} J_{2\perp}\vec{S}_i \cdot \vec{S}_k \quad (1)$$

where J is the intralayer NN magnetic coupling parament, $J_{1\perp}$ and $J_{2\perp}$ represent the interlayer NN and 2NN magnetic coupling paraments, respectively. Four magnetic configurations shown in Figure S11 for AA-stacking and AB-stacking bilayer $GdGe_2$ have been calculated and their energies are described as equation (2) and (3), respectively.

$$E_{FM}^{FM} = E_0 - 24J \cdot |S|^2 - 4J_{1\perp} \cdot |S|^2 - 24J_{2\perp} \cdot |S|^2$$

$$E_{FM}^{AFM} = E_0 - 24J \cdot |S|^2 + 4J_{1\perp} \cdot |S|^2 + 24J_{2\perp} \cdot |S|^2 \quad (2)$$

$$E_{AFM}^{FM} = E_0 + 8J \cdot |S|^2 - 4J_{1\perp} \cdot |S|^2 + 8J_{2\perp} \cdot |S|^2$$

$$E_{AFM}^{AFM} = E_0 + 8J \cdot |S|^2 + 4J_{1\perp} \cdot |S|^2 - 8J_{2\perp} \cdot |S|^2$$

$$E_{FM}^{FM} = E_0 - 24J \cdot |S|^2 - 12J'_{1\perp} \cdot |S|^2 - 12J'_{2\perp} \cdot |S|^2$$

$$E_{FM}^{AFM} = E_0 - 24J \cdot |S|^2 + 12J'_{1\perp} \cdot |S|^2 + 12J'_{2\perp} \cdot |S|^2 \quad (3)$$

$$E_{AFM}^{FM} = E_0 + 8J \cdot |S|^2 + 4J'_{1\perp} \cdot |S|^2 - 12J'_{2\perp} \cdot |S|^2$$

$$E_{AFM}^{AFM} = E_0 + 8J \cdot |S|^2 - 4J'_{1\perp} \cdot |S|^2 + 12J'_{2\perp} \cdot |S|^2$$

E_0 means the ground state energy of nonmagnetic state, superscript and subscript represent the type of interlayer and intralayer magnetic couplings, respectively. S is the spin vector of magnetic atoms. The calculated magnetic coupling paraments were summarized in Table S1.

Table S1. Magnetic ground state, magnetic coupling paraments of intralayer and interlayer exchange interactions in AA-stacking and AB-stacking bilayer $GdGe_2$. J is the magnetic coupling parament between intralayer nearest neighboring (NN) Gd atoms, $J_{1\perp}$ and $J_{2\perp}$ represent the magnetic coupling paraments between the interlayer NN and second NN (2NN) Gd atoms, respectively.

	Ground state	J (meV)	$J_{1\perp}$(meV)	$J_{2\perp}$(meV)
AA-stacking	AFM	0.389	-0.054	-0.022
AB-stacking	FM	1.041	0.495	0.198

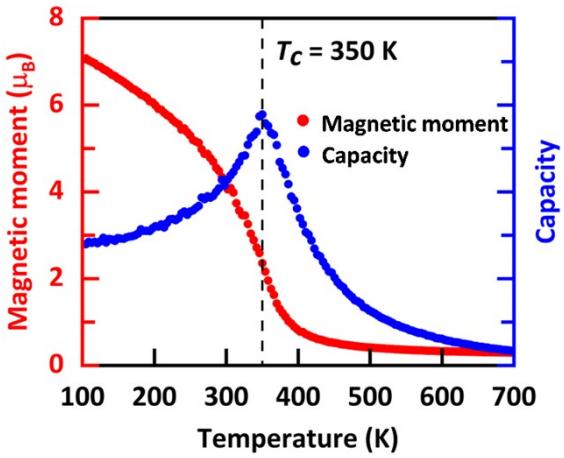

Figure S14. The evolution of magnetic moment of Gd atom (red) and specific heat (blue) with respect to temperature for AB-stacking bilayer $GdGe_2$.