# Supporting Information

## Theoretical Study of the Magnetic Mechanism of a Pca21 $C_4N_3$ Monolayer and the Regulation of Its Magnetism by Gas Adsorption

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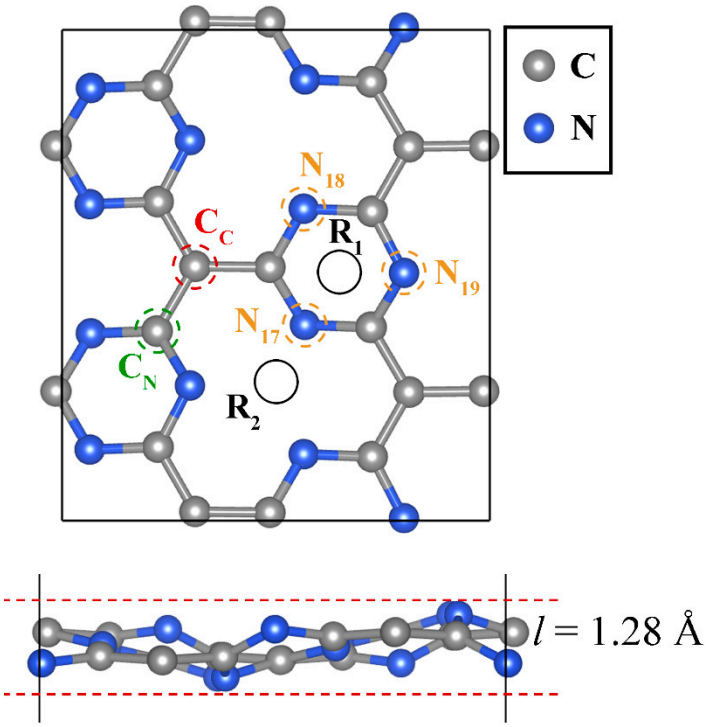



**Figure S1.** Top and side views of the Pca21 $C_4N_3$ monolayer. Blue spheres represent N atoms, and gray spheres indicate C atoms. The red dashed circle indicates $C_C$ atoms, while the green dashed circle represents $C_N$ atoms. The five adsorption sites: **N**, **$C_N$**, **$C_C$**, **R1**, and **R2** are the top sites above the N, $C_N$, $C_C$, R1, and R2, respectively. The orange dashed circles indicate three N atoms (designated as $N_{17}$, $N_{18}$, and $N_{19}$) in the 6-membered microcycles of the Pca21 $C_4N_3$.

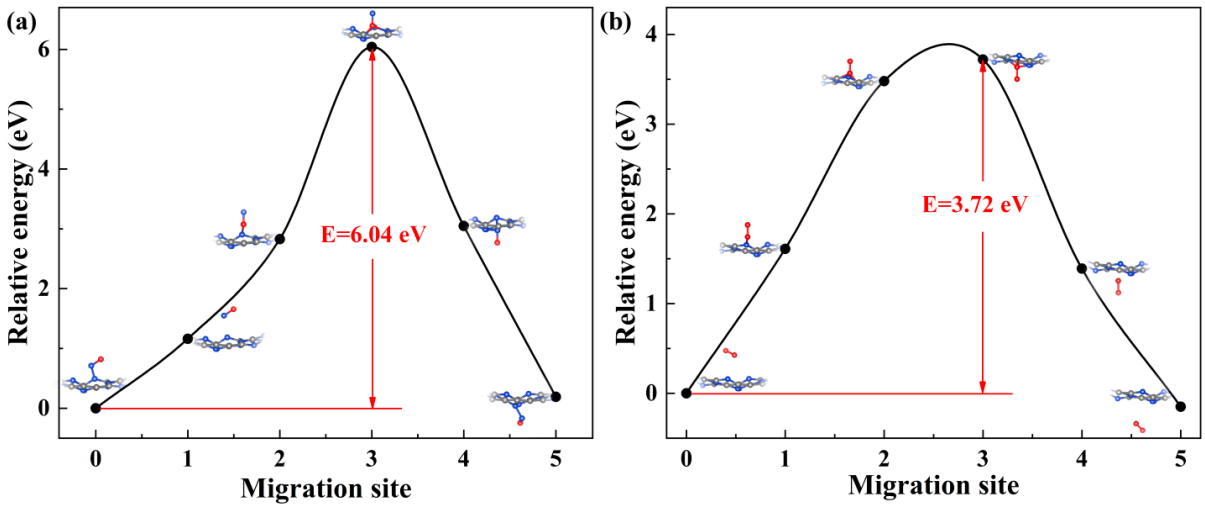



**Figure S2.** The energy barriers for (a) NO and (b) $O_2$ diffusion in the 12-membered macrocycles of a Pca21 $C_4N_3$ monolayer.

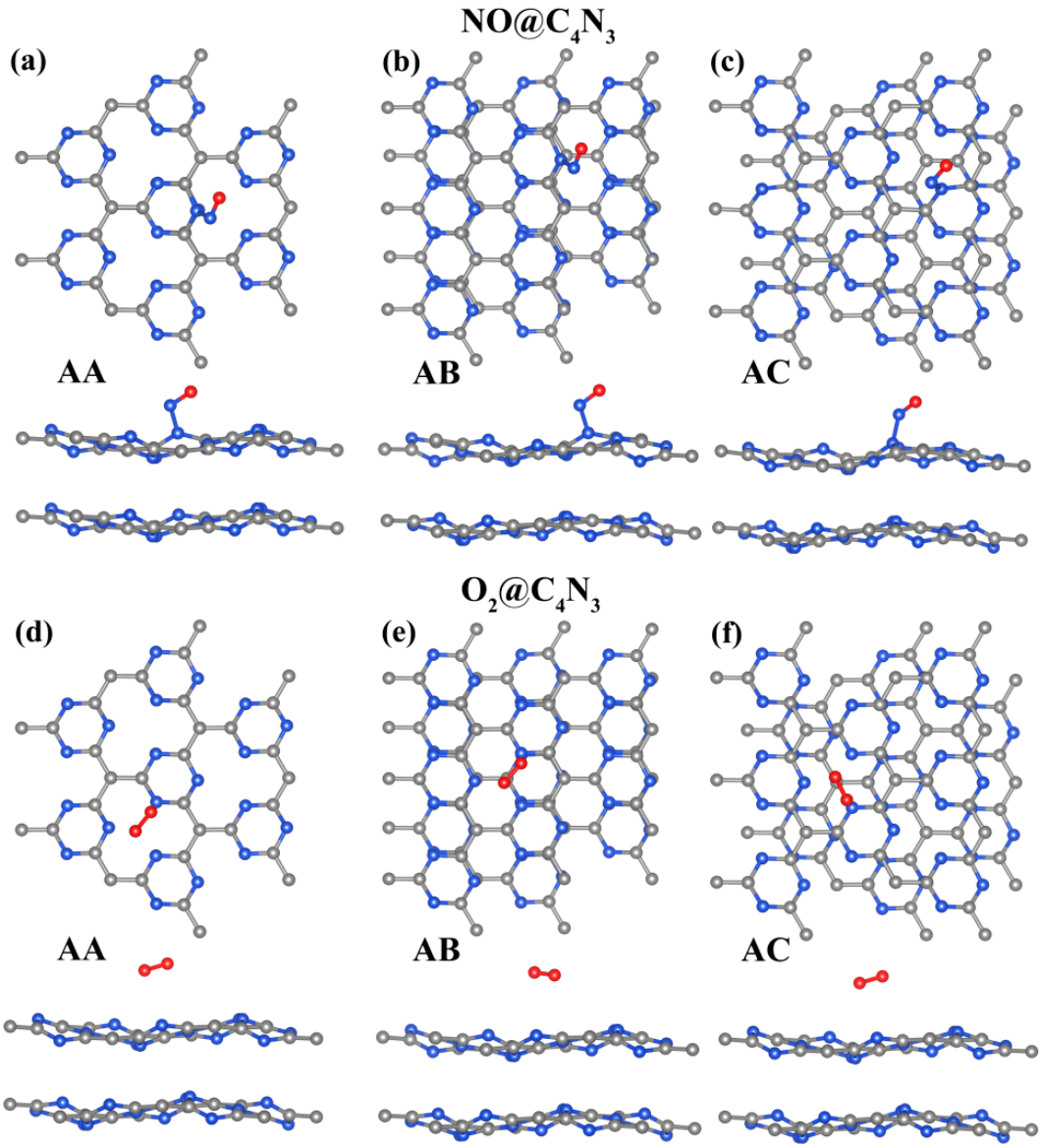



**Figure S3.** The top (upper) and side (lower) views of NO adsorbed on (a) AA, (b) AB, and (c) AC stacking patterns, and $O_2$ adsorbed on (d) AA, (e) AB, and (f) AC stacking patterns for the Pca21 $C_4N_3$ bilayer. AA stacking pattern: the C and N atoms of the upper layer recombined with the C and N atoms of the lower layer, respectively. AB stacking pattern: the C and N atoms of the upper layer are overlapped with the N and C atoms of the lower layer, i.e., the AB stacking pattern is shifted by one C-C bond distance with respect to the AA stacking pattern. AC stacking pattern: the C and N atoms of the upper layer are coincident with those of the lower layer, i.e., the AC stacking pattern is shifted by one C-N bond distance relative to the AB stacking pattern.

**Table S1**. The net magnetic moment (*MM*) and adsorption energy ($E_{ads}$) for NO and $O_2$ adsorbed on $C_4N_3$ monolayer and bilayer with different stacking patterns.

| Pca21 $C_4N_3$ | monolayer | AA | AB | AC |
|---|---|---|---|---|
| $E_{ads}$ (eV) for NO adsorbed systems | -1.14 | -1.26 | -0.85 | -1.07 |
| $E_{ads}$ (eV) for $O_2$ adsorbed systems | -0.11 | 0.66 | -0.11 | -0.44 |
| *MM* ($\mu_B$) for pure systems | 4.00 | 8.00 | 8.00 | 8.00 |
| *MM* ($\mu_B$) for NO adsorbed systems | 2.99 | 7.00 | 6.97 | 7.00 |
| *MM* ($\mu_B$) for $O_2$ adsorbed systems | 6.00 | 10.00 | 10.00 | 10.00 |

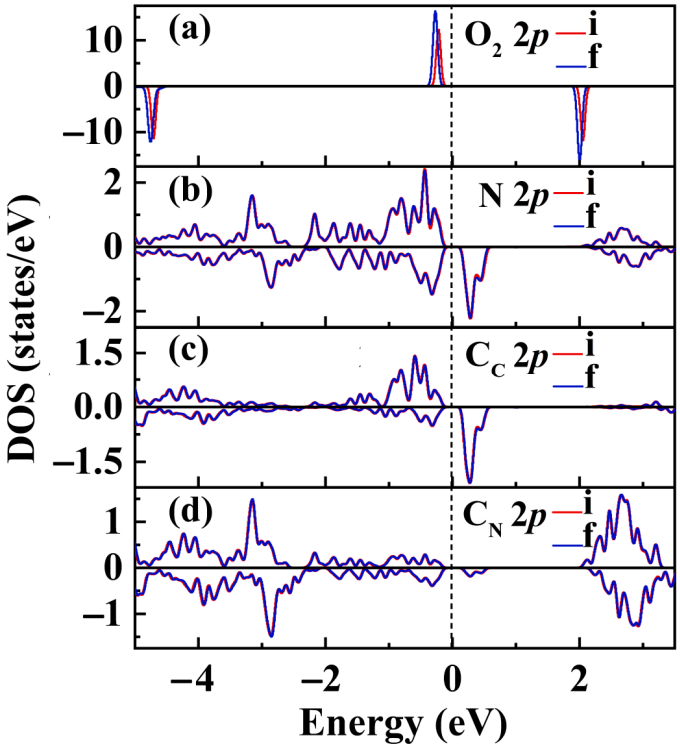



**Figure S4.** The spin-resolved PDOS of 2*p* for (a) $O_2$, and (b) three N, (c) three $C_C$ and (d) six $C_N$ in the 12-membered microcycles at the **R2** adsorption site of the Pca21 $C_4N_3$. The red lines labeled with **i** indicate the 2*p* states before $O_2$ adsorption, while the blue lines labeled with **f** represent the 2*p* states after $O_2$ adsorption.