

Supporting Information for “Tunable Half-metallic Magnetism in Atom-thin Holey Two-dimensional C₂N Monolayer”

Sai Gong,[†] Wenhui Wan,[†] Shan Guan,^{‡,†} Bo Tai,[‡] Chang Liu,[†] Botao Fu,[†]
Shengyuan A. Yang,^{*,‡} and Yugui Yao^{*,†}

*Beijing Key Laboratory of Nanophotonics and Ultrafine Optoelectronic Systems, School of
Physics, Beijing Institute of Technology, Beijing 100081, China, and Research Laboratory for
Quantum Materials, Singapore University of Technology and Design, Singapore 487372,
Singapore*

E-mail: shengyuan_yang@sutd.edu.sg; ygyao@bit.edu.cn

*To whom correspondence should be addressed

[†]Beijing Key Laboratory of Nanophotonics and Ultrafine Optoelectronic Systems, School of Physics, Beijing Institute of Technology, Beijing 100081, China

[‡]Research Laboratory for Quantum Materials, Singapore University of Technology and Design, Singapore 487372, Singapore

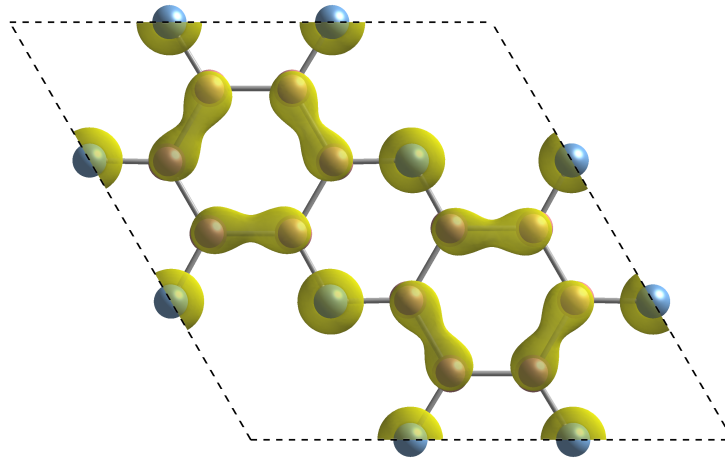


Figure 1: Spatial distribution of the spin polarization for the doping density $6 \times 10^{13}/\text{cm}^2$. The polarization appears on both C and N atomic sites.

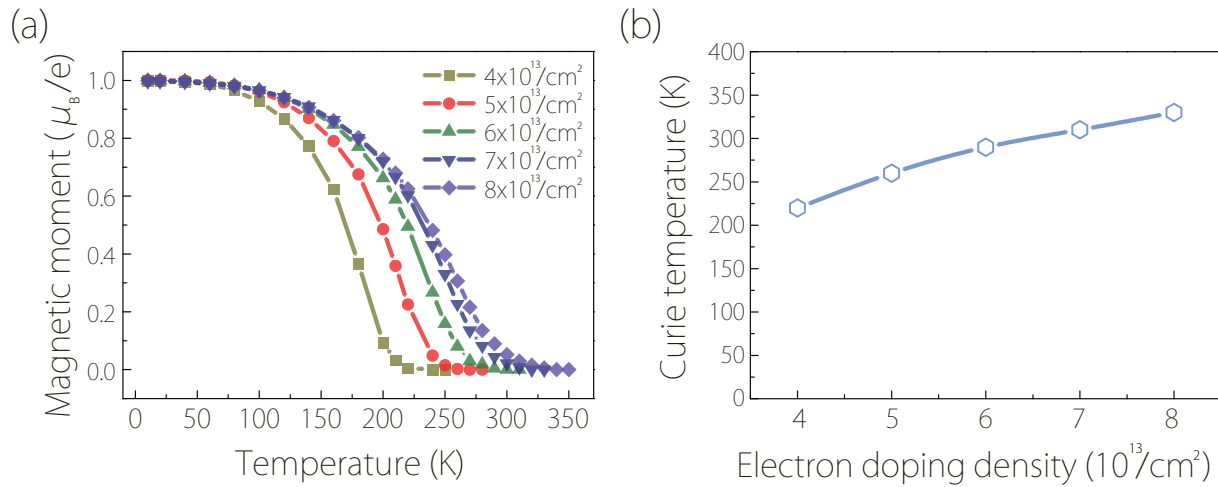


Figure 2: (a) Magnetic moment per carrier versus temperature for several different electron doping densities. Here the magnetic moment is evaluated from the electronic free energy. From the curves, one can estimate the magnetic transition (Curie) temperature. In (b), we plot the estimated Curie temperature against the doping density.

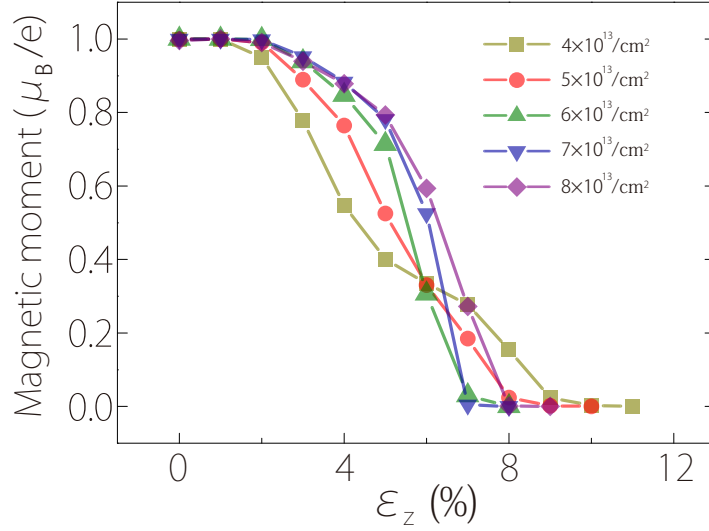


Figure 3: Magnetic moment per carrier versus strain ε_z for several different doping densities. Here the strain ε_z is applied along the zigzag direction (as indicated in Fig. 1(a) in the main text). One observes that the magnetism can be effectively tuned by strain.

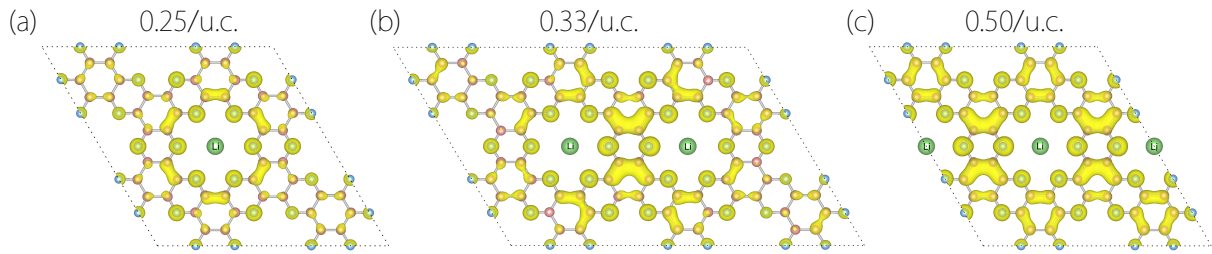


Figure 4: Electron doping to $h2D-C_2N$ by adsorption of Li atoms. The three sub-figures are simulating the doping concentrations of (a) 0.25 e/u.c., (b) 0.33 e/u.c., and (c) 0.50 e/u.c. respectively, where u.c. stands for primitive unit cell. At low doping density, Li tends to stay at the center of the hole, within the same 2D plane as the material. Our calculation confirms that the system indeed become ferromagnetic. The plot in this figure shows the spin density distribution (i.e. the difference between spin-up and spin-down densities). The results for Na and K adsorption are similar.

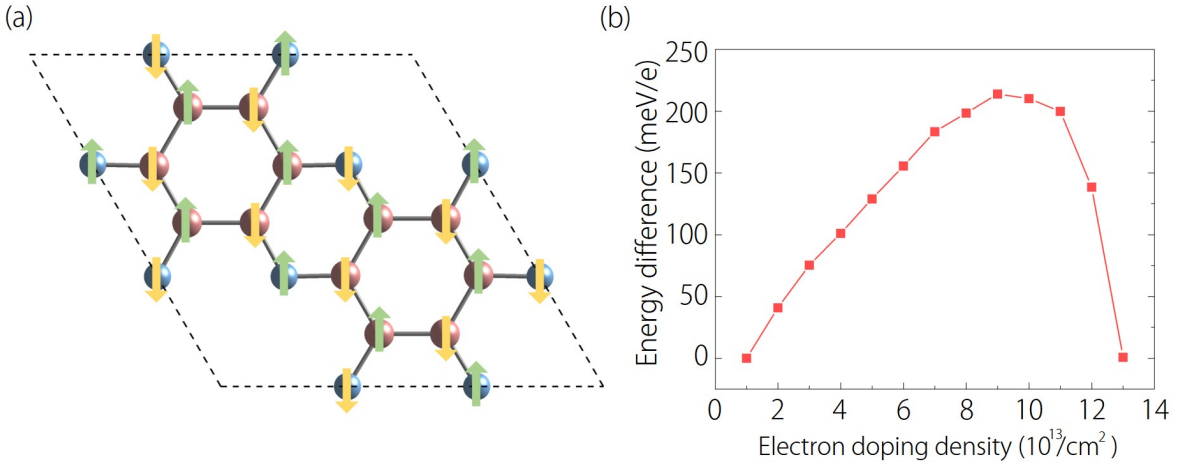


Figure 5: (a) Antiferromagnetic configuration. (b) Energy difference ($E_{AFM} - E_{FM}$) between AFM and FM configurations as a function of doping.

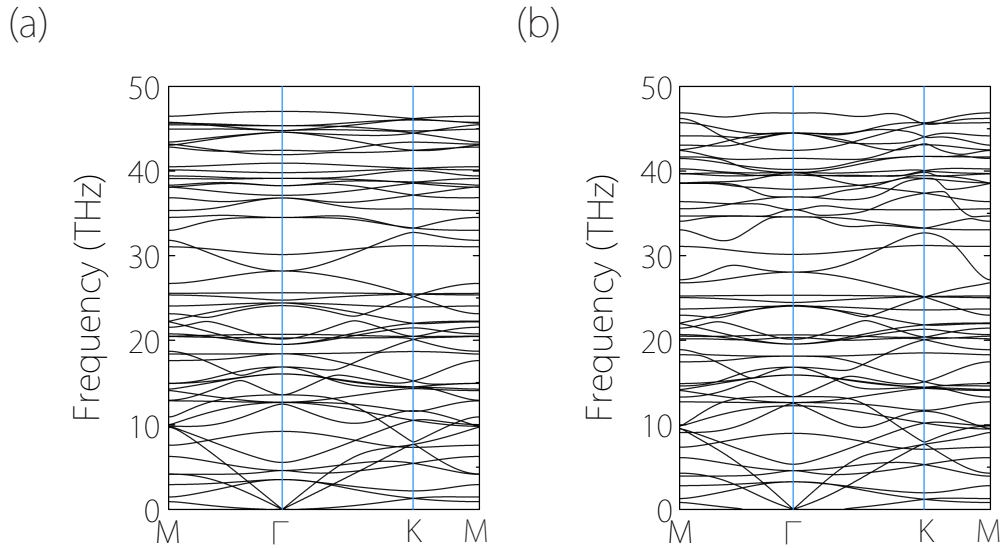


Figure 6: Phonon spectra for (a) undoped C₂N, and (b) C₂N with doping density of $6 \times 10^{13}/\text{cm}^2$.

Table 1: Electron doping to $h2D-C_2N$ by adsorption of alkali atoms: N denotes the doping density, E_b is the binding energies of the adsorbed atoms, and M is resulting magnetic moment per electron carrier.

Adatom	N (e/u.c.)	E_b (eV/adatom)	M (μ_B/e)
Li	0.25	4.194	0.9309
	0.33	4.220	0.8300
	0.50	4.295	0.7974
Na	0.25	4.106	0.9765
	0.33	4.141	0.9173
	0.50	4.167	0.8501
K	0.25	4.087	0.9998
	0.33	4.117	0.9800
	0.50	4.142	0.9544