Electronic supplementary information (ESI)

Ultralow diffusion barrier induced by intercalation in layered N-based cathode materials for Sodium-ion batteries

Yundan Jiang, Wangping Xu, Wei Zhao*, Juexian Cao*

Department of Physics & Hunan Institute of Advanced Sensing and Information Technology, Xiangtan University, Xiangtan 411105, PR China

*Corresponding authors: zhaowei@xtu.edu.cn; jxcao@xtu.edu.cn

Details of setting the magnetic order and the U value

Since the 3d transition metal Cr is magnetic, spin polarization is taken into account in all calculations. In order to obtain the magnetic ground state of the system, a series of ferromagnetic and antiferromagnetic states are calculated. The antiferromagnetic states include Neel type, Stripe type, and Zigzag type, as shown in Fig. S1.[1] The results show that the antiferromagnetic state with Neel type is the ground state of the system.

PBE+U can accurately describe the energy correction induced by strongly correlated d electrons in transition metal systems. Based on the linear correspondence method[2], the value of U is determined according to formula (S1):

$$U = x^{-1} - x_0^{-1} \approx \left(\frac{\partial N_I^{SCF}}{\partial \varphi_I} \right)^{-1} - \left(\frac{\partial N_I^{NSCF}}{\partial \varphi_I} \right)^{-1} \quad \text{(S1)}$$

Where N^{SCF} and N^{NSCF} are the numbers of d electrons under the self-consistent field and non-self-consistent field respectively. φ is the additional potential. (Here x=1.35, x_0=0.23, as shown in Fig. S2).

Note: Ref. 2 was cited in the main article as Ref. 31.

1 X.-J. Dong, J.-Y. You, B. Gu and G. Su, *Phys. Rev. Appl.*, 2019, **12**, 014020.

2 M. Cococcioni and S. de Gironcoli, *Phys. Rev. B*, 2005, **71**, 035105.

Supplementary Figures

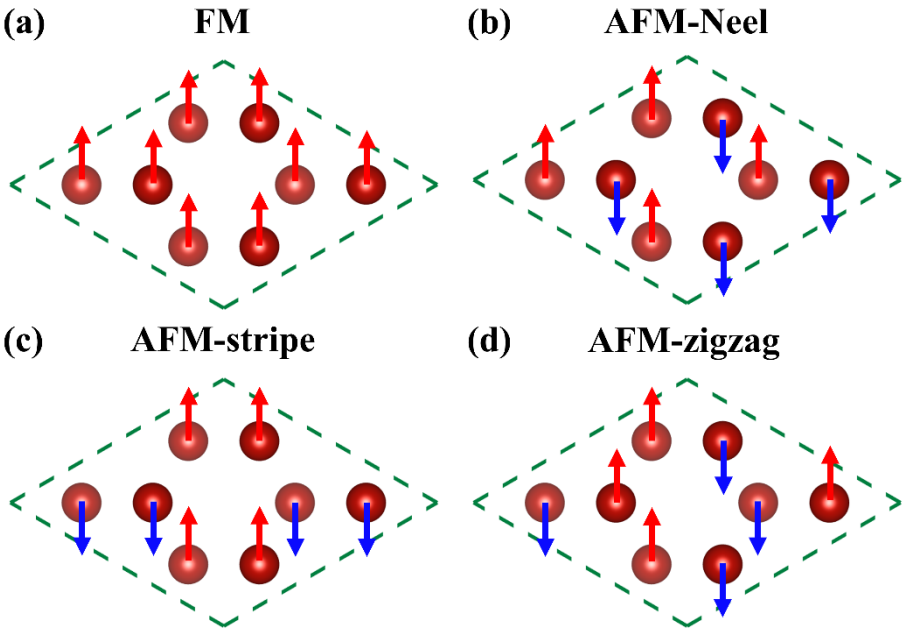

Fig. S1 The four magnetic orders of the calculated system. (The dark red ball represents Cr, and the red and blue arrows represent spin up and spin down respectively)

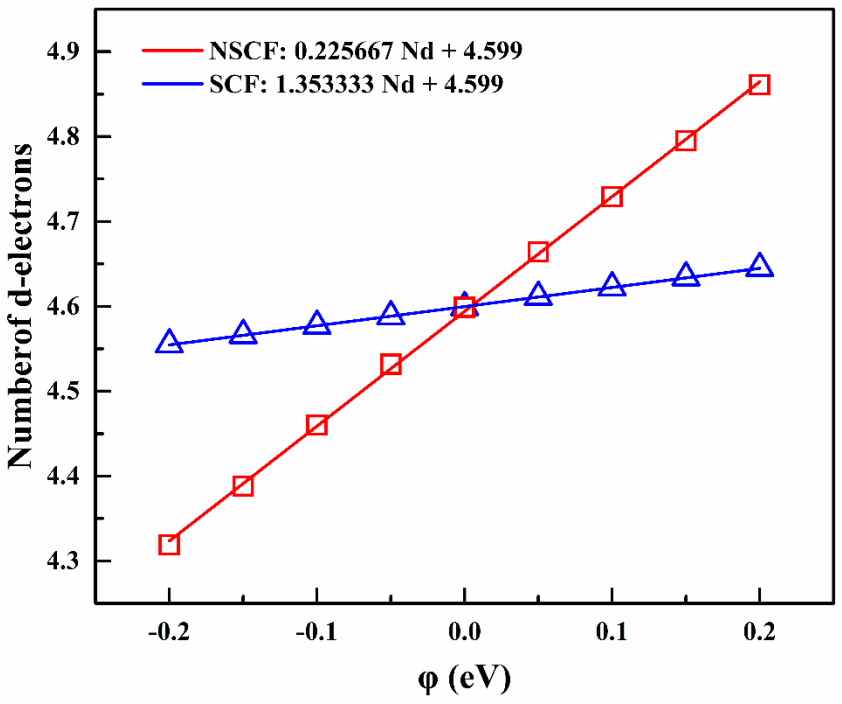

Fig. S2 A linear fit of the number of d-electrons on Cr as a function of the additional potential φ.

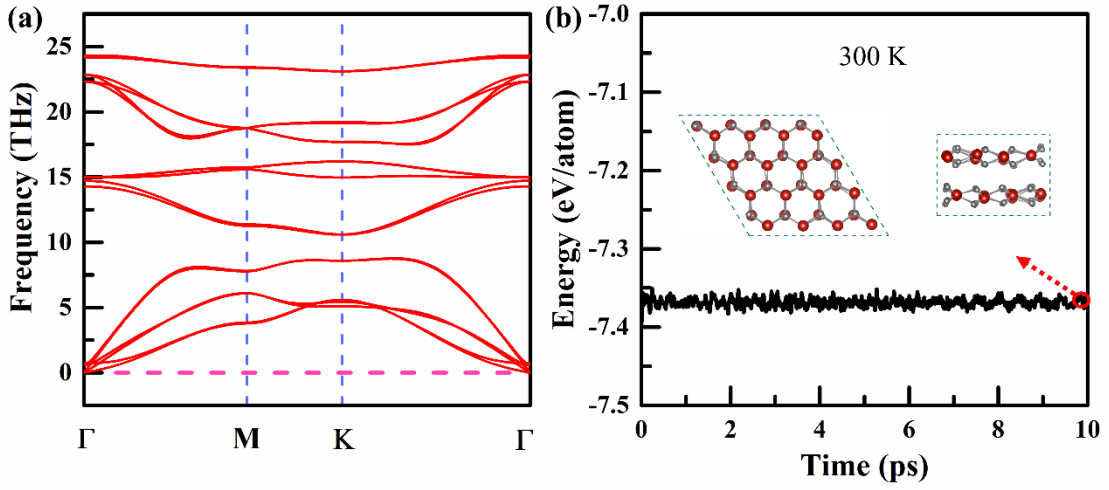

Fig. S3 (a) Phonon spectrum of 2H-phase CrN_2. (b) The total energy per atom fluctuations of 2H-pahse CrN_2 in AIMD calculations using a canonical ensemble (NVT) at a temperature of 300 K. Inserts show the averaged structures in the last 100 fs times in AIMD calculations.

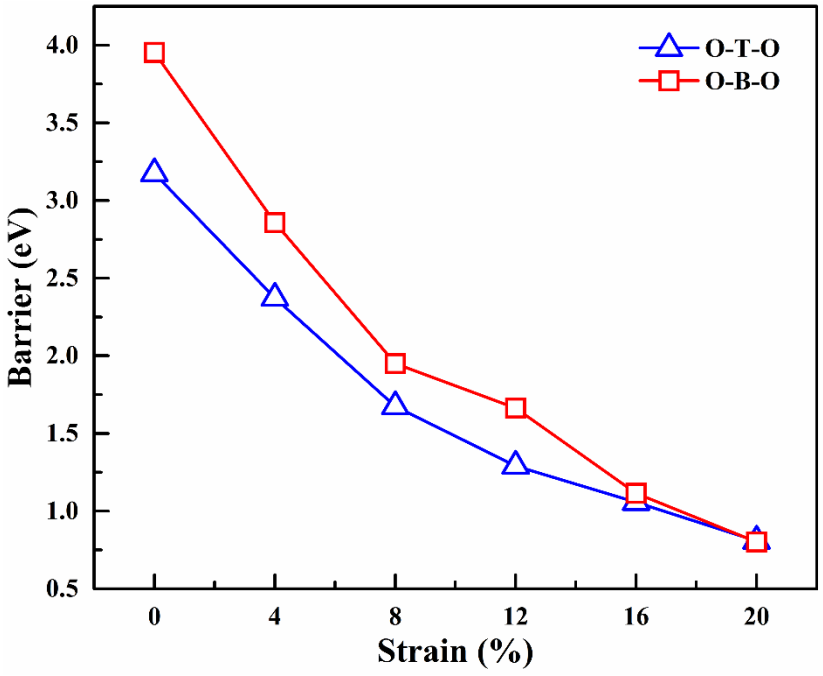

Fig. S4 The relationship between the potential barrier and c-axis strain.

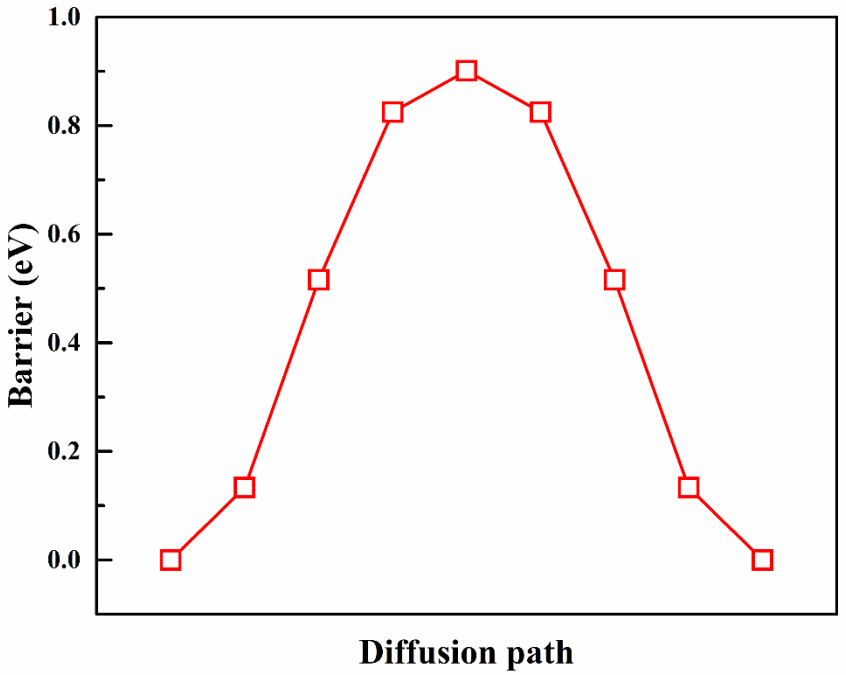

Fig. S5 The energy profile of sodium ions along the diffusion path in the CrN_2 without intercalation.

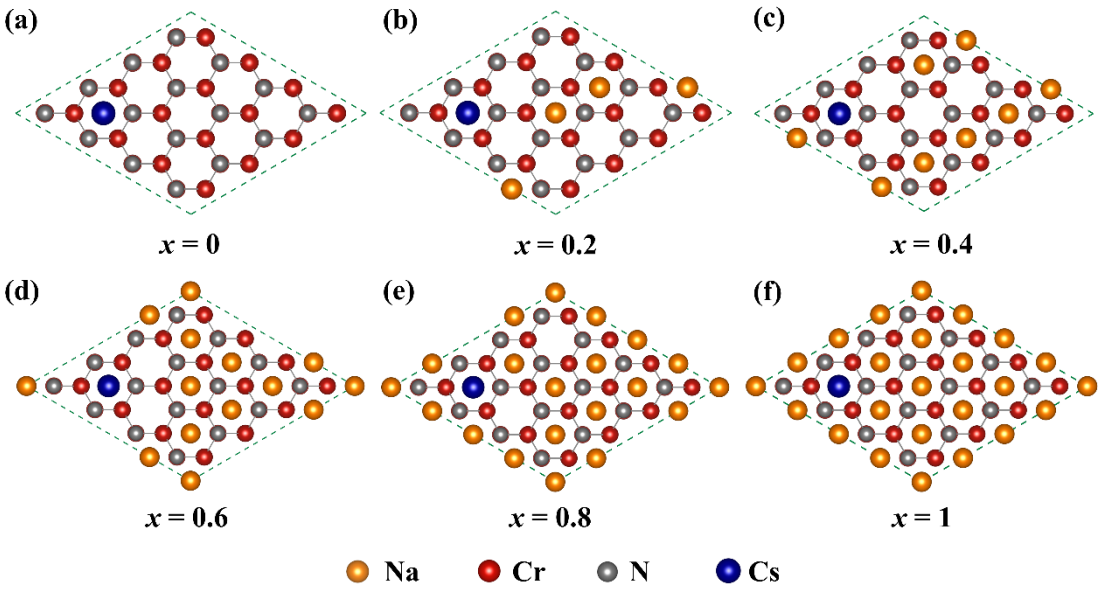

Fig. S6 The most stable configuration of $Na_xCs_{1/16}CrN_2$ (x = 0 ~ 1) under different sodium concentrations. (Orange, dark blue, dark red, and gray spheres represent Na, Cs, Cr, and N atoms, respectively.).

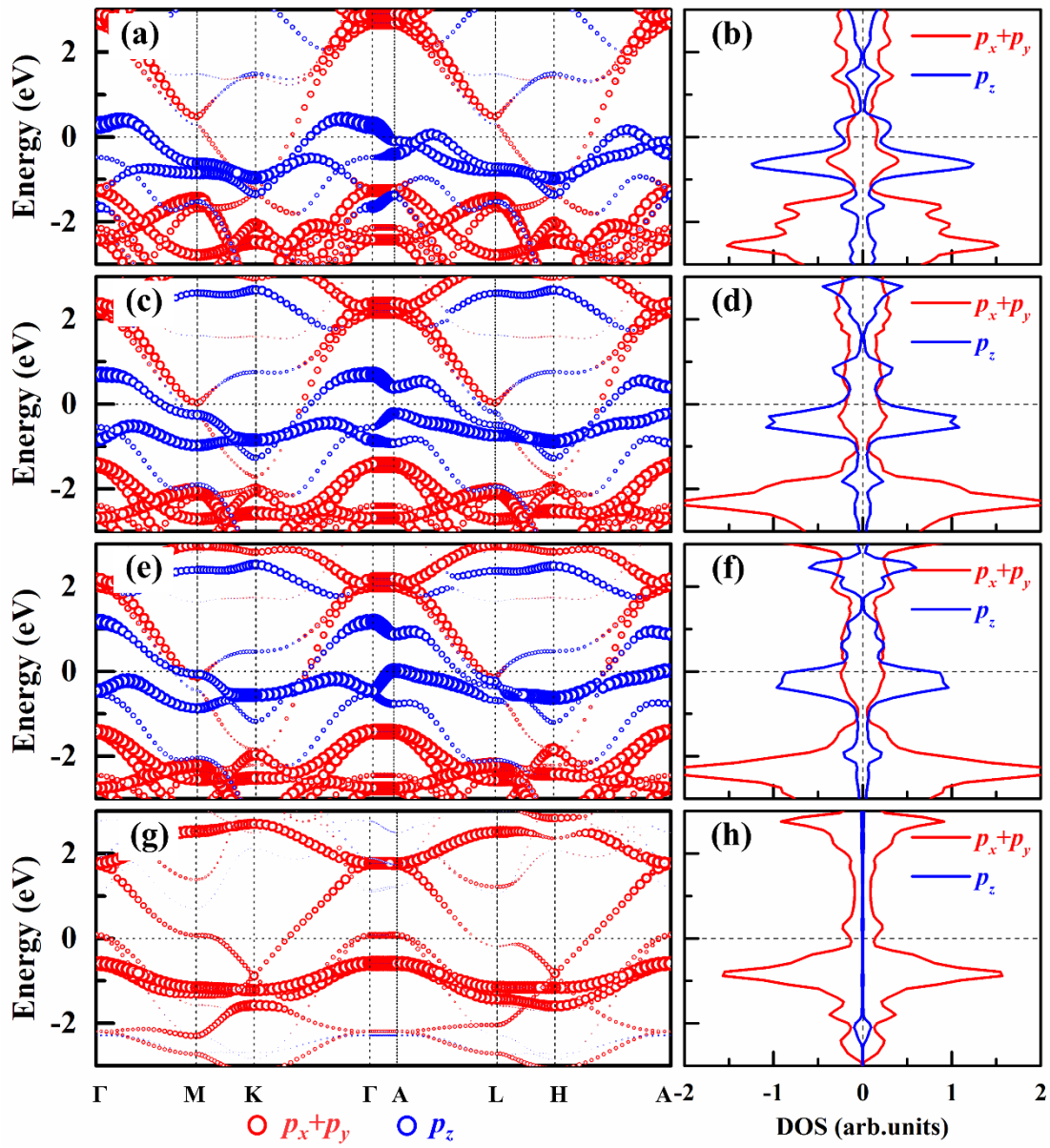

Fig. S7 Calculated projected band (PBAND) and projected density of states (PDOS) for N in $NaCrN_2$ (a and b), $KCrN_2$ (c and d), $RbCrN_2$ (e and f) and $CsCrN_2$ (g and h), respectively.