Supplementary materials for

# Persistent exchange splitting in a chiral helimagnet $Cr_{1/3}NbS_2$

Na Qin[1*], Cheng Chen[2*], Shiqiao Du[1*], Xian Du[1], Xin Zhang[3], Zhongxu Yin[1], Jingsong Zhou[1], Runzhe Xu[1], Xu Gu[1], Qinqin Zhang[1], Wenxuan Zhao[1], Yidian Li[1], Sung-Kwan Mo[2], Zhongkai Liu[3,4], Shilei Zhang[3,4], Yanfeng Guo[3,4], Peizhe Tang[5,6], Yulin Chen[1,3,4,7‡], and Lexian Yang[1,8‡]

[1]*State Key Laboratory of Low Dimensional Quantum Physics, Department of Physics, Tsinghua University, Beijing 100084, China.*

[2]*Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*

[3]*School of Physical Science and Technology, ShanghaiTech University and CAS-Shanghai Science Research Center, Shanghai 201210, China.*

[4]*ShanghaiTech Laboratory for Topological Physics, Shanghai 200031, China.*

[5]*School of Materials Science and Engineering, Beihang University, Beijing 100191, China.*

[6]*Max Planck Institute for the Structure and Dynamics of Matter, Center for Free Electron Laser Science, 22761 Hamburg, Germany.*

[7]*Department of Physics, Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, UK.*

[8]*Frontier Science Center for Quantum Information, Beijing 100084, China.*

[*]*These authors contribute equally to this work.*

[‡]*Email address: LXY: lxyang@tsinghua.edu.cn, YLC: yulin.chen@physics.ox.ac.uk*

**This file contains the following sections:**

**I. Details about the fit of EDCs and MDCs;**

**II. Comparison between the experimental and calculated band structures near the Fermi level;**

**III. Calculated band structure of $Cr_{1/3}NbS_2$ with different Coulomb interaction between Cr atoms.**

## S I Details of EDC fit and MDC fit for $Cr_{1/3}NbS_2$

In order to extract the exchange splitting between the $\beta_1$ and $\beta_2$ bands, we fit the momentum distribution curves (MDCs) and energy distribution curves (EDCs) to Lorentzians. Figure S1(a) shows ARPES spectra of $Cr_{1/3}NbS_2$ taken at 15 K, the same as Fig. 5(a) in the main text. Figure S1(b) shows the fit of the MDC at the Fermi level ($E_F$) to three Loretzians representing the three band crossings $E_F$. The yellow line is a linear background. Figure S1(c) shows the fit of EDC at the Fermi momentum of $\beta_2$ ($k_x$ = 0.31 Å$^{-1}$). The band splitting between the $\beta_1$ and $\beta_2$ bands can be reliably extracted from the data fit of either the EDC or MDC.

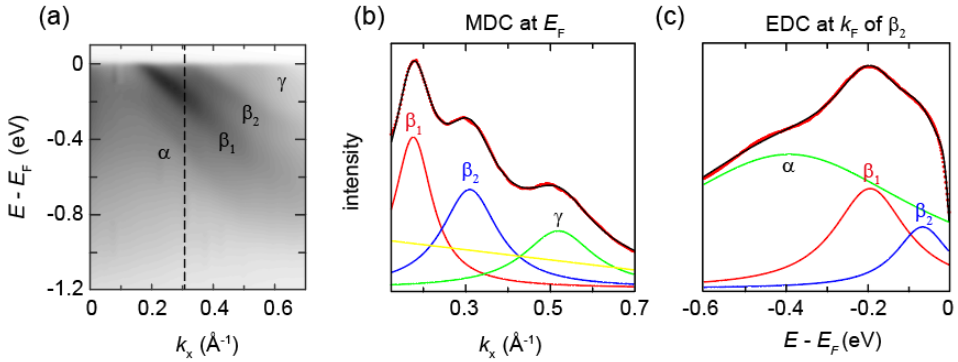


**Fig. S1.** (a) ARPES spectra of $Cr_{1/3}NbS_2$ measured at 15 K. (b) The fit of MDC at $E_F$ to three Lorentzians. The yellow line is a linear background. (c) The fit of EDC at the Fermi momentum ($k_F$) of the $\beta_2$ band.

## S II. Comparison between the experimental and calculated band structures near the Fermi level;

Both the measured and calculated band structures of $Cr_{1/3}NbS_2$ in the main text are in good agreement with previous reports. However, due to the electronic correlation between Cr *d* electrons and complicated magnetic ordering in the system, the calculated result is much more complicated than the measured band structure. The deviation between the experiment and calculation remains a puzzle in the electronic of $Cr_{1/3}NbS_2$.

Figure S2 compares the measured and calculated band structure of $Cr_{1/3}NbS_2$. In the paramagnetic (PM) state at 155 K, we observe a broad feature, corresponding to the calculated Nb $d_{x2-y2}/d_{xy}$ bands [Fig. S2(b)]. In the FM states, there are complicated band reconstruction due to the exchange

splitting in both experiment and calculation. However, in the calculation, the band structure is more complicated. There are not only more bands crossing $E_F$ but also two less dispersive bands below $E_F$ in the calculation that are absent in our work and previous experiments. Further experimental and theoretical investigations are highly demanded to understand the deviations in Fig. S2.

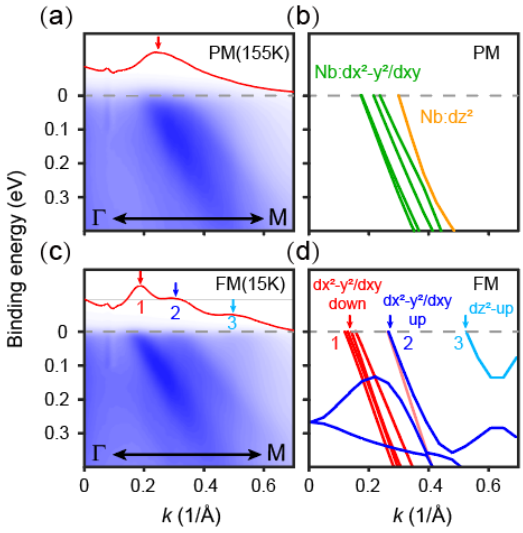


**Fig S2.** Comparison between the experimental (a, c) and calculated (b, d) band structures of $Cr_{1/3}NbS_2$ in the paramagnetic (PM) (a, b) and ferromagnetic (FM) (c, d) states.

## S III. Calculated band structure of $Cr_{1/3}NbS_2$ with different Coulomb interaction between Cr atoms.

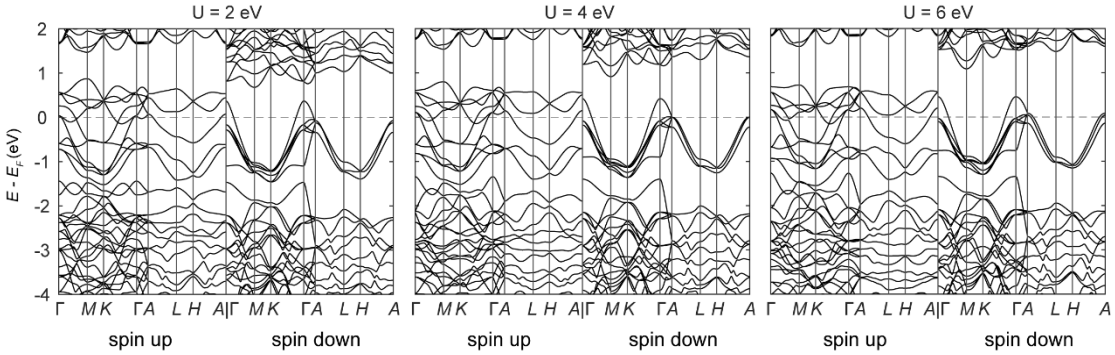


**Fig S3.** Calculated band structures of $Cr_{1/3}NbS_2$ with U = 2 eV, 4 eV, 6 eV, respectively.

In Fig. 4 of the main text, we used an onsite Coulomb interaction of U = 4 eV in the calculation of the band structure of $Cr_{1/3}NbS_2$, which is the typical value used for Cr *d* states in many compounds [1-3]. Figure S3 shows the calculated band structure of $Cr_{1/3}NbS_2$ for U = 2 eV, 4

eV, 6 eV. The Coulomb interaction mainly changes the band gap and dispersions in high binding energies, while the bands crossing $E_F$ are barely changed.

**Reference**


[1] S. Shi, A. L. Wysocki, and K. D. Belashchenko, Phys. Rev. B 79, 104404 (2009).

[2] E. Şaşıoğlu, C. Friedrich, and S. Blügel, Phys. Rev. B 83, 121101 (2011).

[3] L. Vaugier, H. Jiang, and S. Biermann, Phys. Rev. B 86, 165105 (2012).