

Supporting information

Colossal Magnetization and Giant Coercivity in Ion Implanted (Nb, Co) MoS₂ Crystals

Sohail Ahmed^t, Xiang-Yuan Carl Cui^ξ, Xiang Ding^{†‡}, Peter Paul Murmu[§], Nina Bao^{#,5}, Xun Geng^t, Shibo Xi[‡], Rong Liu[‡], John Kennedy[§], Tom Wu^t, Lan Wang^o, Kiyonori Suzuki^Ⅴ, Jun Ding⁵, Xueze Chu^{*10}, Sathish Russellraj Clastirusselraj Indirathankam^{*10}, Mingli Peng[◇], Ajayan Vinu^{*10}, Simon Peter Ringer^{ξ*} and Jiabao Yi^{0*}

[†]. School of Materials Science and Engineering, UNSW, Sydney, New South Wales 2052, Australia

^ξ. Australian Centre for Microscopy & Microanalysis, and School of Aerospace, Mechanical and Mechatronic Engineering, The University of Sydney, New South Wales 2006, Australia

[‡]. School of Energy and Power Engineering, Wuhan University of Technology, Wuhan, Hubei, 430063, China

[§]. National Isotope Centre, GNS Science, P.O. Box 31312, Lower Hutt, 5010, New Zealand

[#]. Department of Materials Science and Engineering, National University of Singapore, 119260, Singapore

[‡]. Institute of Chemical and Engineering Sciences, A*STAR, 1 Pesek Road, Jurong Island, 627833, Singapore

^Ⅴ. SIMS Facility, Office of the Deputy-Vice Chancellor (Research and Development), Western Sydney University, Locked Bag 1797, Penrith, New South Wales 2751, Australia

^o. School of Science, RMIT University, Melbourne, Victoria 3001, Australia

^Ⅴ. Department of Materials Science and Engineering, Monash University, Clayton, Victoria 3800, Australia

[◇]. *Key Laboratory of Synthetic and Natural Functional Molecule Chemistry of the Ministry of Education, College of Chemistry and Materials Science, Northwest University, Xi'an 710069, China*

⁰. Global Innovative Centre for Advanced Nanomaterials, School of Engineering, The University of Newcastle, Callaghan, New South Wales 2308, Australia

Email: jiabao.yi@newcastle.edu.au; simon.ringer@sydney.edu.au

Table S1. Magnetization and coercivity of MoS₂ based diluted magnetic semiconductor

No	Sr	Material	Saturation Magnetization (emu/cm ³)	Coercivity	Ref
1	Re doped MoS ₂		0.0076	170 Oe	[1]
2	F adsorbed MoS ₂		1.25	< 400 Oe	[2]
3	Cu doped MoS ₂		0.115	~ 100 Oe	[3]
4	V doped MoS ₂		0.335	1870 Oe	[4]
5	Co doped MoS ₂		4	400 Oe	[5]
6	Ni doped MoS ₂		3	175 Oe	[5]
7	Mn doped MoS ₂		0.075	1076 Oe	[6]
8	Nb and/or Co doped MoS ₂		1800	9000 Oe	This work

[1]. Kochat, V. Apte, A. Hachtel, J.A. Kumazoe, H. Krishnamoorthy, A. Susarla, S. Idrobo, J.C. Shimojo, F. Vashishta, P. Kalia, R. Nakano, A. Tiwary, C.S. Ajayan, P.M. Adv. Mater., 2017, 29, 1703754.

[2]. Gao, D. Shi, S. Tao, K. Xia, B. Xue, D. Nanoscale, 2015, 7, 4211.

[3]. Xia, B. Guo, Q. Gao, D. Shi, S. Tao, K., J. Phys. D: Appl. Phys., 2016, 49, 165003.

[4] Ahmed, S. Ding, X. Bao, N. Bian, P. Zheng, R. Wang, Y. Murmu, P.P. Kennedy, J.V. Liu, R. Fan, H. Suzuki, K. Ding, J. Yi, J., Chem. Mater., 2017, 29, 9066.

[5] Martinez, L.M. Delgado, J.A. Saiz, C.L. Cosio, A. Wu, Y. Villagrán, D. Gandha, K. Karthik, C. Nlebedim, I.C. Singamaneni, S.R., J. Appl. Phys., 2018, 124, 153903.

[6]. Wang, J. Sun, F. Yang, S. Li, Y. Zhao, C. Xu, M. Zhang, Y. Zeng, H., Appl. Phys. Lett., 2016, 109, 092401.

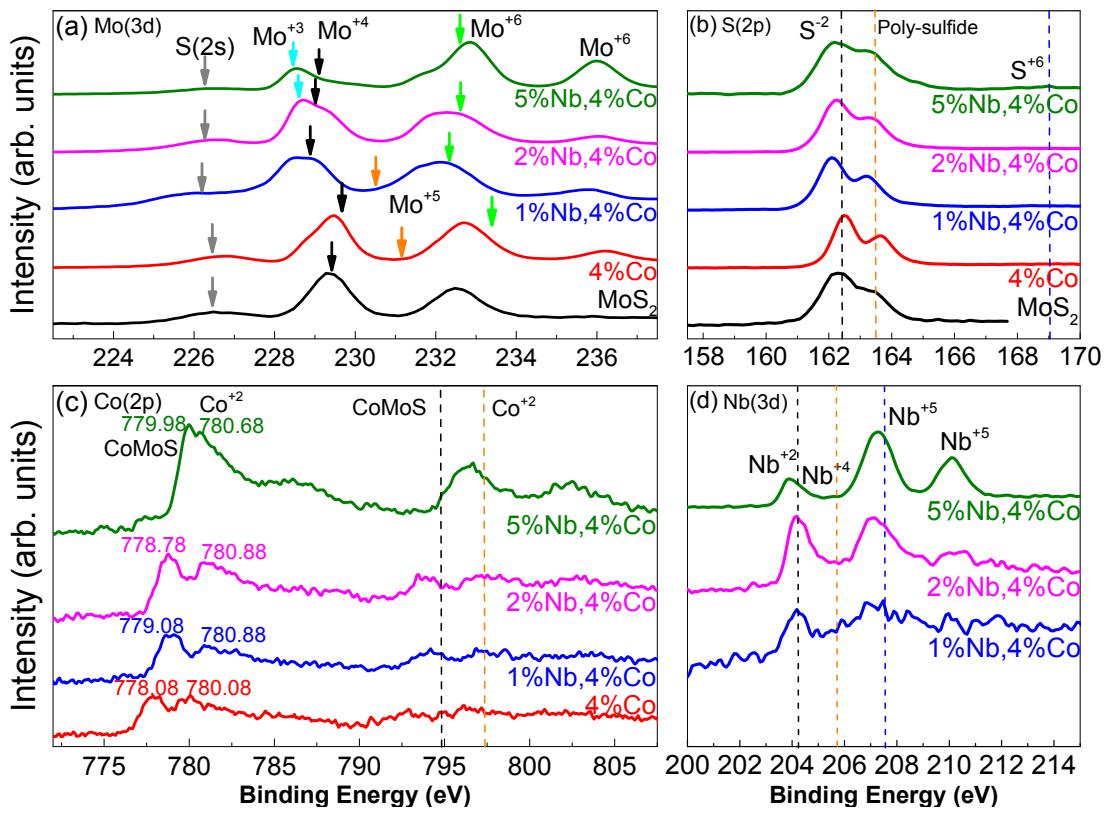


Figure S1. XPS spectra of Co & Nb co-doped MoS₂ with (a) Mo(3d), (b) S(2p), (c) Co(2p) and (d) Nb(3d) core levels.

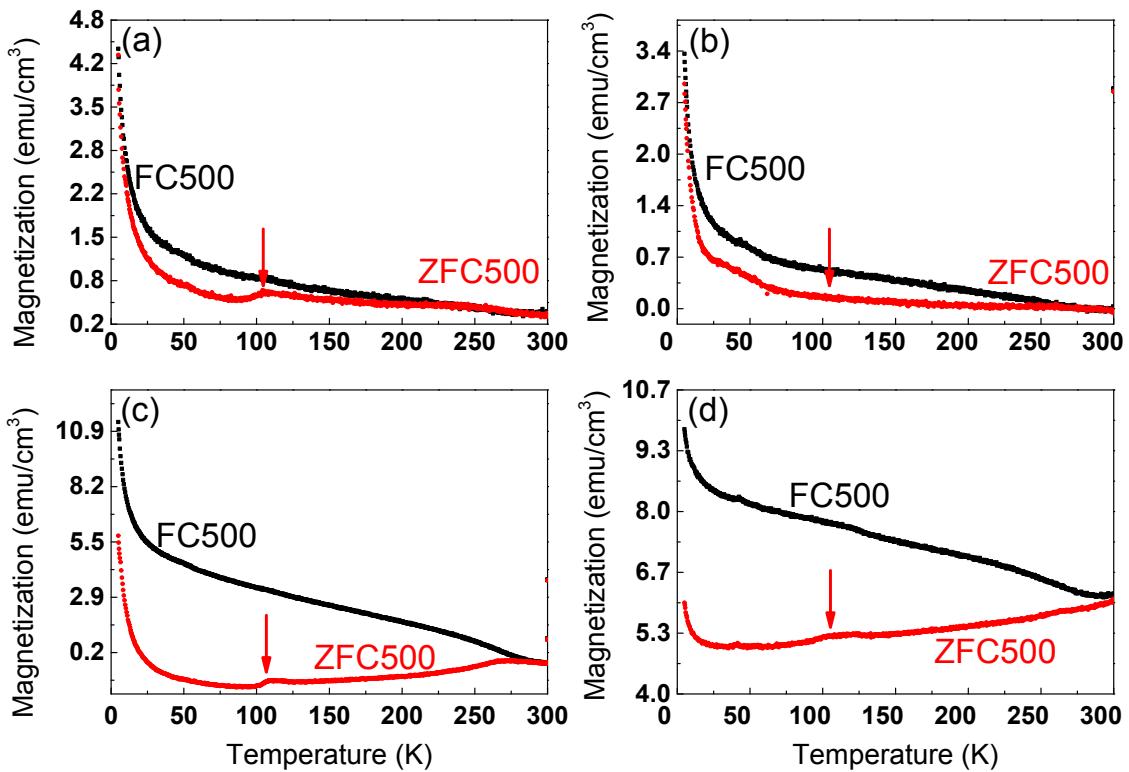


Figure S2. Zero field cooling (ZFC) and field cooling (FC) of (a) 4 at% Co doped MoS₂; (b) 1 at% Nb + 4 at% Co co-doped MoS₂; (c) 2 at% Nb + 4 at% Co co-doped MoS₂ and (d) 5 at% Nb + 4 at% Co co-doped MoS₂.

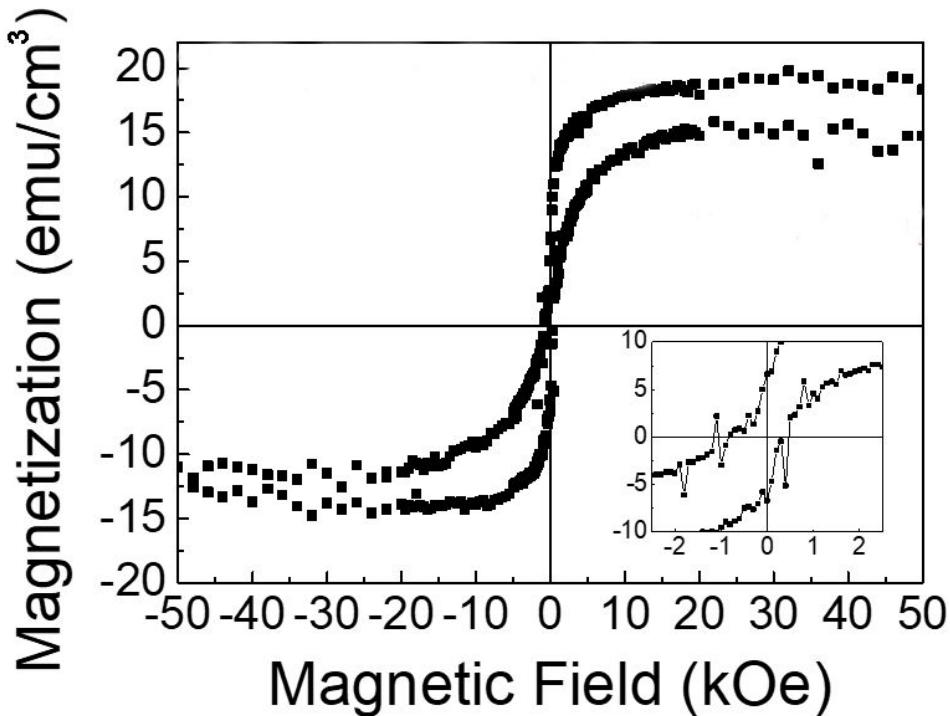


Figure S3. The hysteresis loop of Annealed 2 at% Nb + 4 at% Co co-doped MoS₂ at 100 K shows ferromagnetic signal. The magnetization at the applied magnetic field of 50 kOe at 100 K is enhanced a little (18.351 emu/cm³) compared to that of un-annealed 2 at% Nb + 4 at% Co co-doped MoS₂ (16.14 emu/cm³). Moreover, a reduction in coercivity (822.9 Oe) is observed in the sample of annealed 2 at% Nb + 4 at% Co co-doped MoS₂ (inset). The reduction in coercivity may be attributed to the removal of defects and lattice strain induced during ion implantation process.

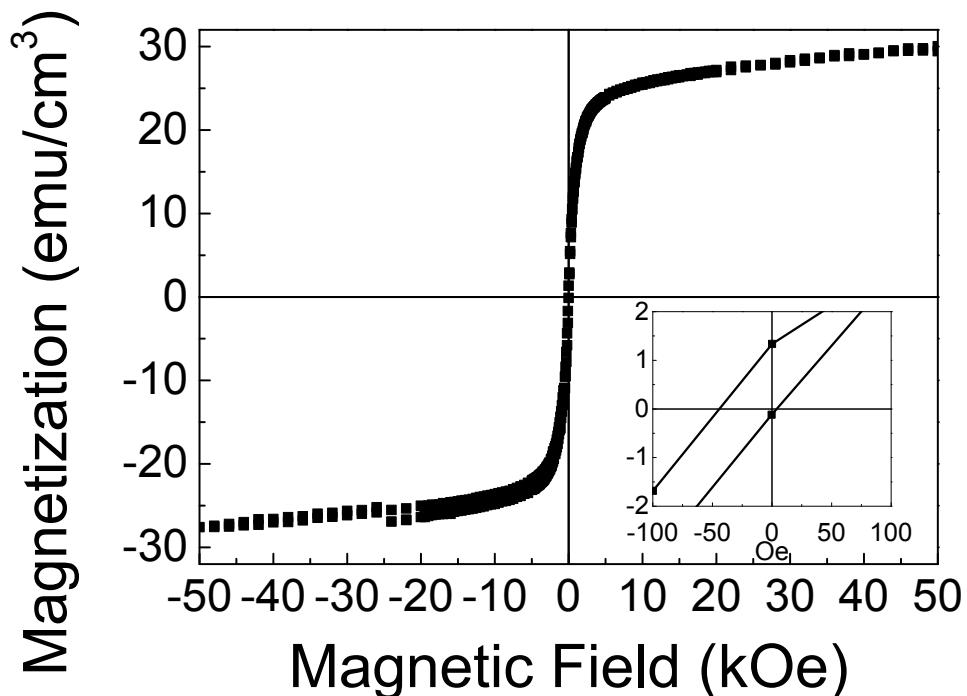


Figure S4. The hysteresis loop of 2 at.% Nb doped MoS₂ at 100 K shows ferromagnetic signal. The magnetization at the applied magnetic field of 50 kOe at 100 K is 30.03 emu/cm³. Moreover, a reduction in coercivity (44.2 Oe) is observed in the sample of 2 at.% Nb doped MoS₂ (inset).

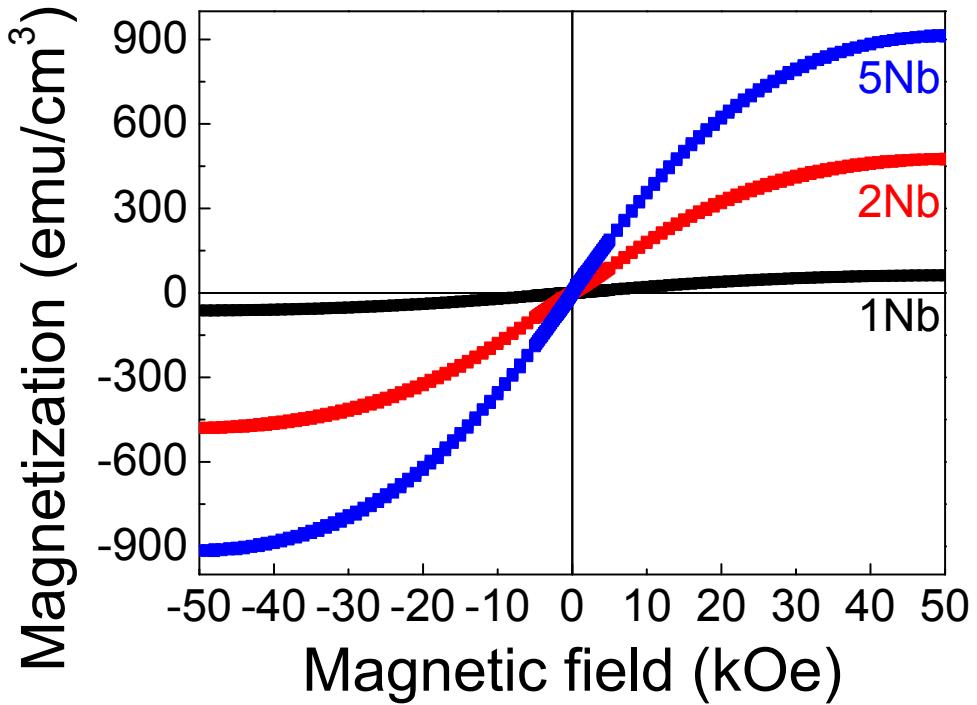


Figure S5. The hysteresis loop of MoS₂ doped with different concentrations annealed at 500 K in Ar environment. At the applied magnetic field of 50 kOe at 5 K, the magnetization of 915 emu/cm³, 473 emu/cm³ and 62.43 emu/cm³ were observed attributed to the 5 at.% Nb, 2 at.% Nb and 1 at.% Nb doped MoS₂.

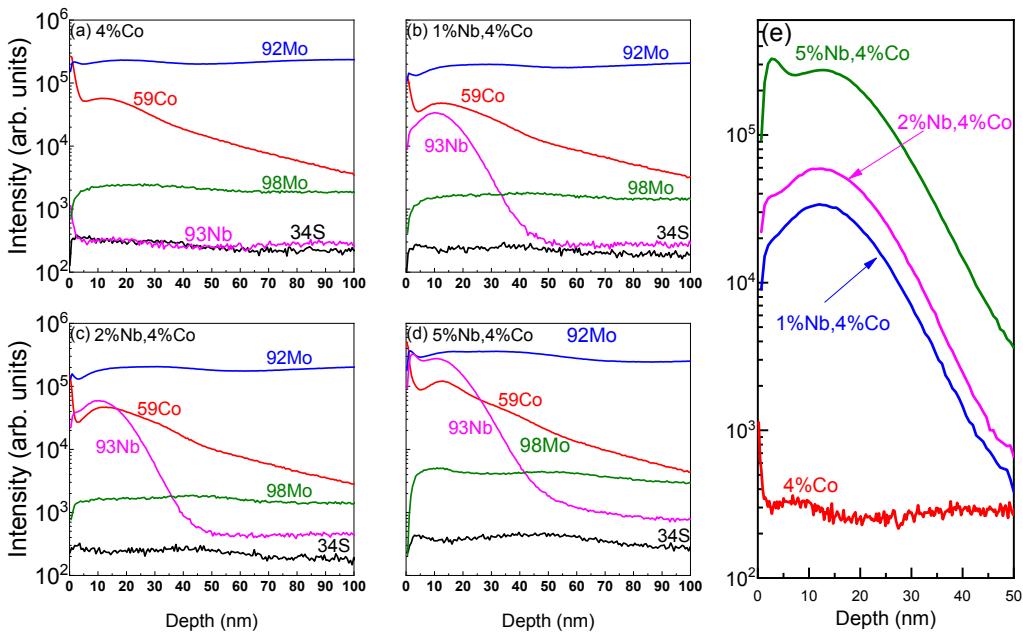


Figure S6. Depth profile of Mo, Co, Nb, and S element in ion implanted single crystals of (a) 4 at.% Co doped MoS₂; (b) 1 at.% Nb + 4 at.% Co co-doped MoS₂; (c) 2 at% Nb + 4 at.% Co co-doped MoS₂ and (d) 5 at% Nb and 4 at.% Co co-doped MoS₂. (e) Depth profile of Nb with different doping concentrations in MoS₂.

Figure S6 shows the concentration of Co and Nb elements as a function of depth for all four Co and Nb-doped MoS₂ samples. In all four samples (Figure S6a-d), Co shows Gaussian profile with the peak at around 12.5 nm from a surface and then a linear reduction in concentration for further depth. Standard deviation (also called straggling) for this set of sample is 7 nm. Therefore, the thickness of the doping is considered around 20 nm.

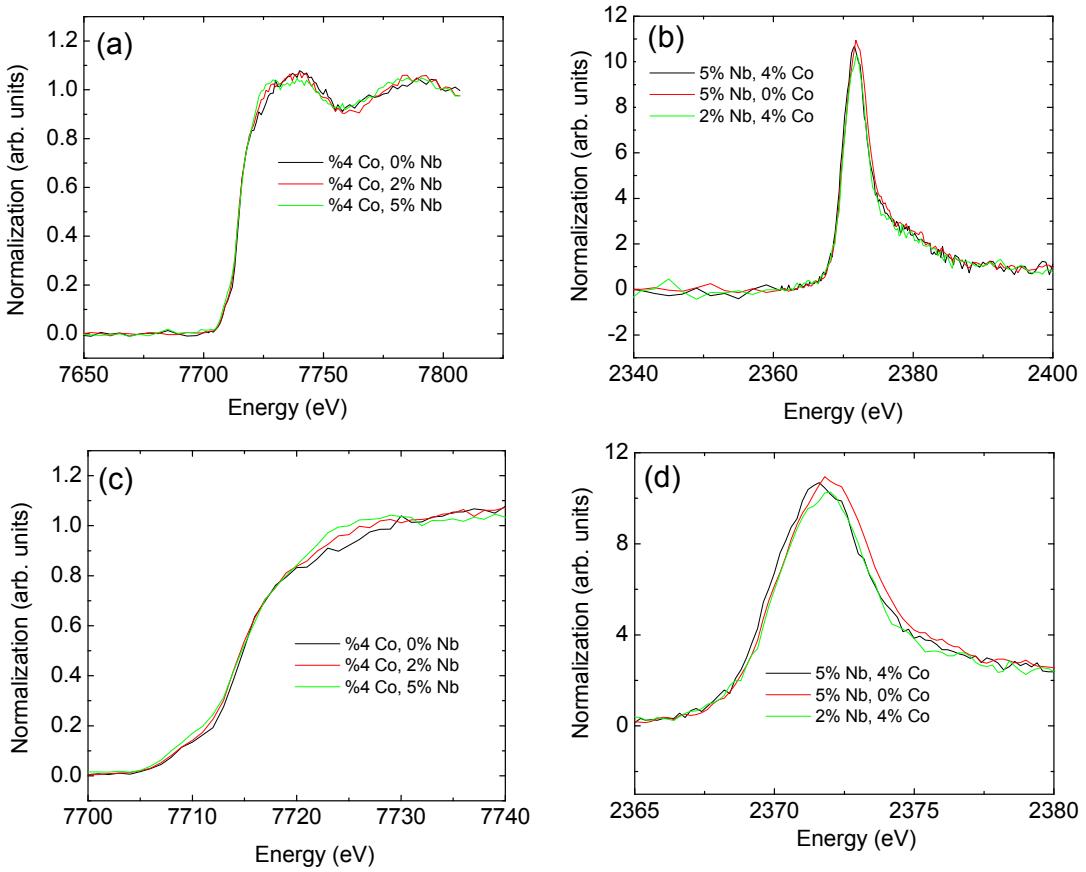


Figure S7: (a) X-ray Absorption Near Edge Structure (XANES) of Co K edge; (b) XANES of Nb L_3 edge; (c) Small scale of (a); (d) Small scale of (b). Due to the bulk signal from XANES, there is no significant difference for different concentration of doping.