Electronic Supplementary Information for the manuscript

Unveiling the origin of room-temperature ferromagnetism in monolayer VSe_2: the role of extrinsic effects

Danil W. Boukhvalov and Antonio Politano

S1. Vibrational experiments on chemisorption on monolayer VS_2

To elucidate the eventual formation of surface vanadium-oxide phases, we have carried out a vibrational investigation by high-resolution electron energy loss spectroscopy (HREELS) on monolayer VSe_2 on graphite, which was prepared using the exact experimental procedures reported in the Experimental methods of the paper by Bonilla et al. [1].

The use of HREELS provides unambiguous fingerprint regarding surface chemical bonds and adsorption at solid surfaces. Specifically, the inspection of the HREELS spectrum is a particularly powerful tool to identify vanadium-oxide phases and, correspondingly, the oxidation status of V atoms. As a matter of fact, VO_2 is characterized by surface phonons at 55 and 95 meV [2]; V_2O_3 by an intense feature at 68 meV [3]; and V_2O_5 by several modes at 45, 74, 105, and 126 meV [4]. However, the vibrational spectrum in Fig. R1 does not exhibit any feature even after exposure to 10^5 L of O_2. This unambiguously excludes the formation of surface vanadium-oxide phases, that, if existing, do not cover more than 1-2% of the surface area (corresponding to the minimal sensitivity of HREELS technique).

Notably, the vibrational spectrum remains featureless even after exposure to 10^5 L of H_2, H_2O and CO. The absence of adsorbate-related vibrational modes proves the chemical inertness of monolayer VSe_2 to most common gases, thus experimentally validating our theoretical model concerning chemical reactivity.

We also recorded the vibrational spectrum for the surface kept one year in air. The spectrum is dominated by C-H bending and stretching vibrations at 171 and 365 meV [5], respectively. This could

arise from hydrocarbon contamination from ambient air [6] adsorbed at Se vacancies of the VSe_2 monolayer.

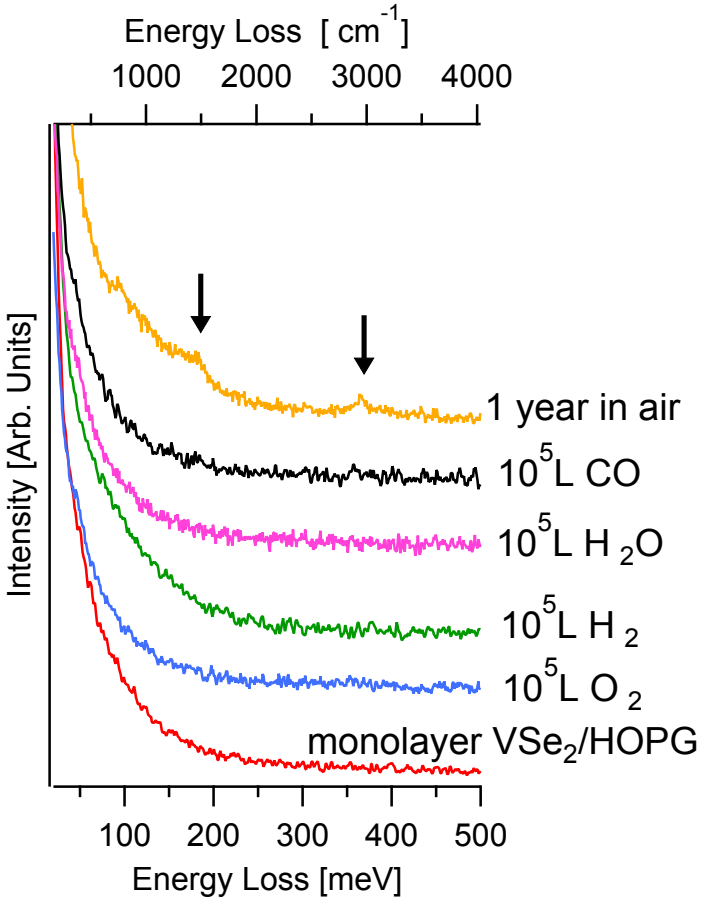

Figure S1: Vibrational spectra for monolayer VSe_2 grown on highly oriented pyrolytic graphite (HOPG) and the same surface modified by the exposure of O_2, H_2, H_2O and CO. The vibrational spectrum for the same surface kept one year in air is also reported.

REFERENCES

1. M. Bonilla, S. Kolekar, Y. Ma, H. C. Diaz, V. Kalappattil, R. Das, T. Eggers, H. R. Gutierrez, M.-H. Phan and M. Batzill, *Nat. Nanotechnol.*, 2018, **13**, 289-293.
2. Z. Chang, S. Piligkos and P. J. Møller, *Phys. Rev. B*, 2001, **64**, 165410.
3. A.-C. Dupuis, M. A. Haija, B. Richter, H. Kuhlenbeck and H.-J. Freund, *Surf. Sci.*, 2003, **539**, 99-112.
4. B. Tepper, B. Richter, A.-C. Dupuis, H. Kuhlenbeck, C. Hucho, P. Schilbe, M. Bin Yarmo and H.-J. Freund, *Surf. Sci.*, 2002, **496**, 64-72.
5. H. Kim, T. Balgar and E. Hasselbrink, *Chem. Phys. Lett.*, 2011, **508**, 1-5.
6. Z. Li, Y. Wang, A. Kozbial, G. Shenoy, F. Zhou, R. McGinley, P. Ireland, B. Morganstein, A. Kunkel, S. P. Surwade, L. Li and H. Liu, *Nat. Mater.*, 2013, **12**, 925-931.