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SUPPLEMENTARY INFORMATION
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Strong room-temperature ferromagnetism in VSe_2 monolayers on van der Waals substrates

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Supplementary Information:

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Supplementary Fig. 1

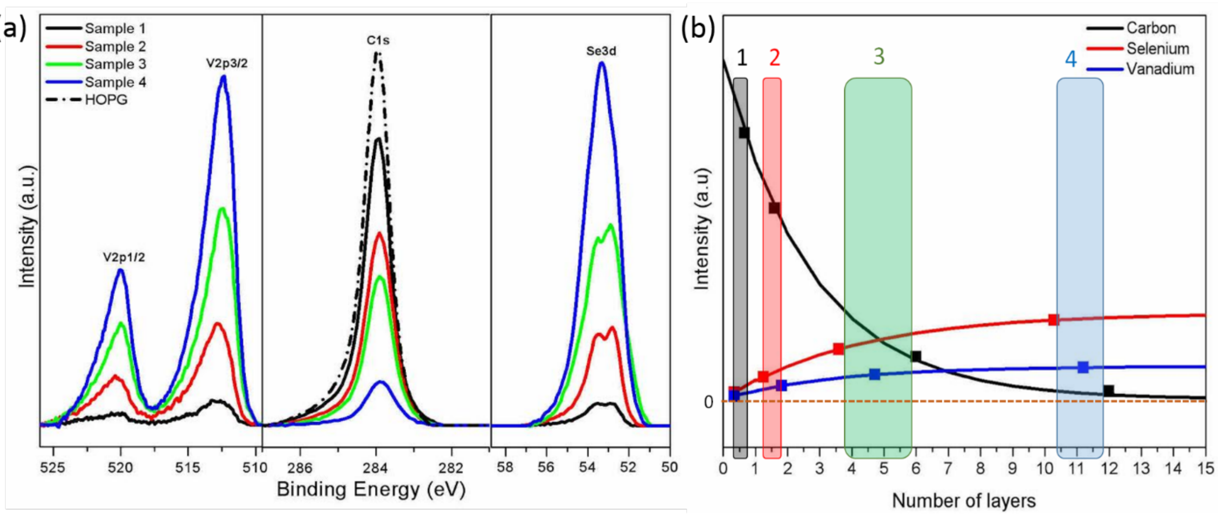

Fig. S1: XPS analysis of VSe2 film growth. XPS data for V-2p, C-1s, and Se-3d core levels for the four films magnetic measurements are discussed are shown in (a). The intensity analysis for estimating the film thickness is shown in (b). The widths of the colored bars represent the estimated uncertainty in the number of layers for these samples.

Supplementary Fig. 2

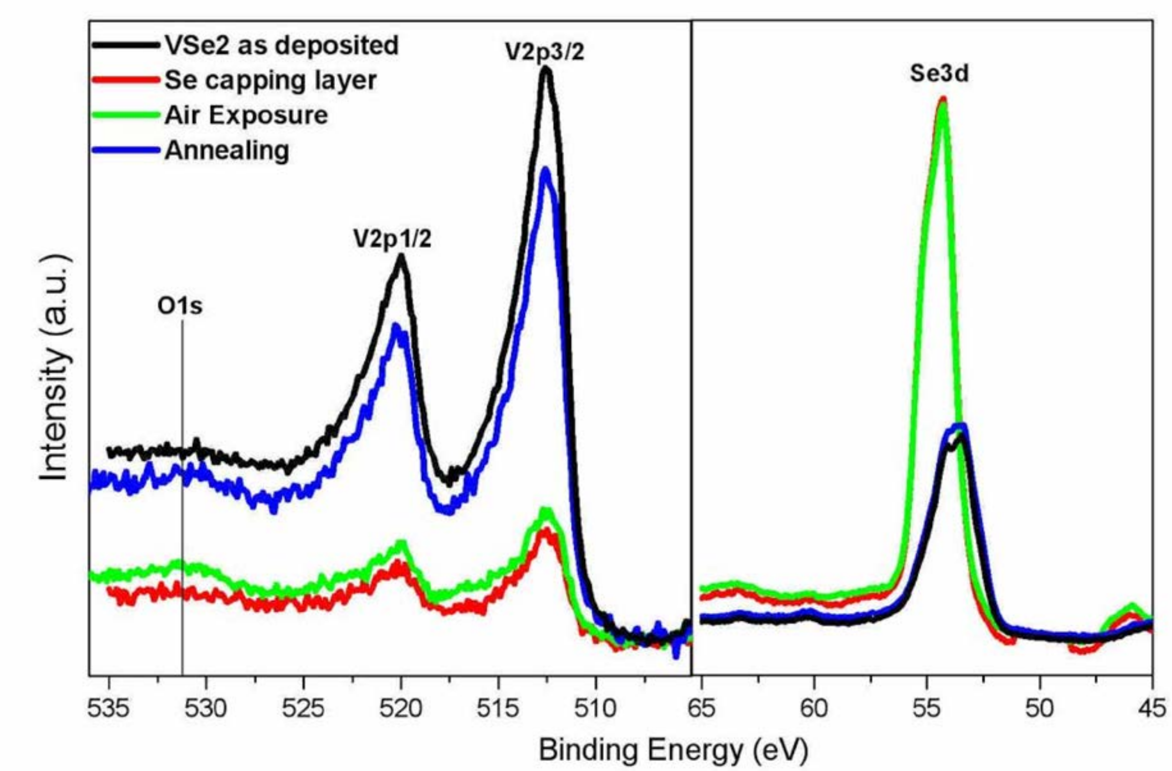

Fig. S2: Air stability of Se-capped VSe_2 ultrathin films. XPS analysis of protection of a VSe_2 film from air exposure with a Se capping layer. V-2p and Se-3d core level peaks are shown for the following sequential sample history: (i) an as-grown film (black), (ii) after capping the film with ~ 10 nm Se layer (red), (iii) exposure to air for 30 h and reintroduced into the vacuum chamber (green), (iv) removal of the Se-capping layer by annealing at 300 °C in UHV (blue). The vanadium and selenium peaks appear almost identical for the as-grown film and after removal of the Se-capping layer, indicating that air exposure does not oxidize or alter the VSe_2 film.

Supplementary Fig. 3

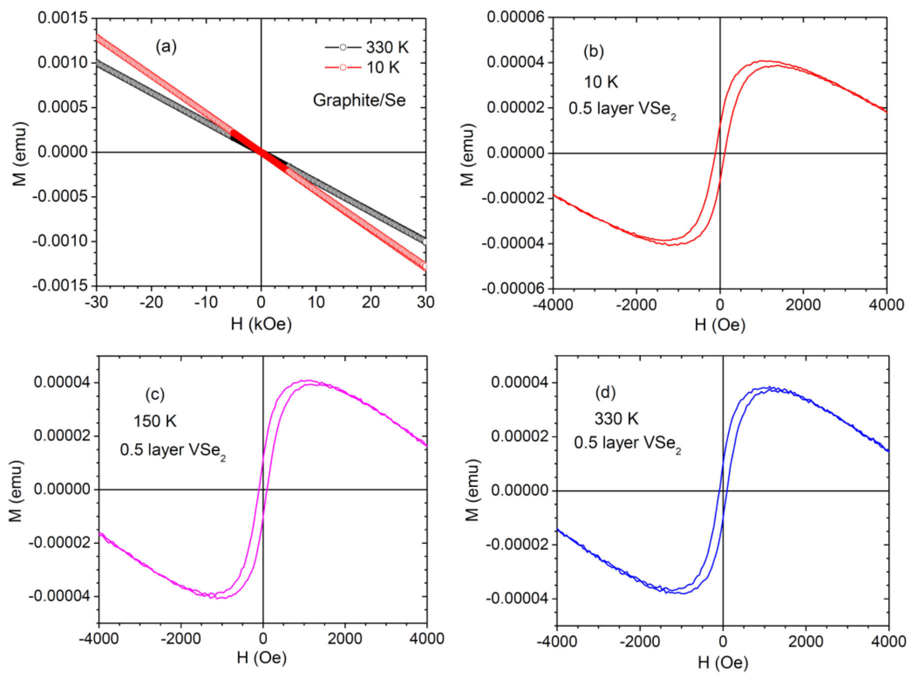

Fig. S3: Diamagnetism of HOPG with Se capping layer (control sample) and intrinsic ferromagnetism of monolayer 1T-VSe_2. (a) Linear field dependence of magnetization M(H) taken at 10 and 330 K indicates a diamagnetic background of the graphite-substrate with Se-capping layer, while well-defined hysteresis loops without subtracting the diamagnetic background of the graphite/Se-capping taken at various temperatures 10 K (b), 150 K (c), and 330 K (d) evidence the strong, intrinsic ferromagnetic ordering above room temperature for monolayer 1T-VSe_2.

Supplementary Fig. 4

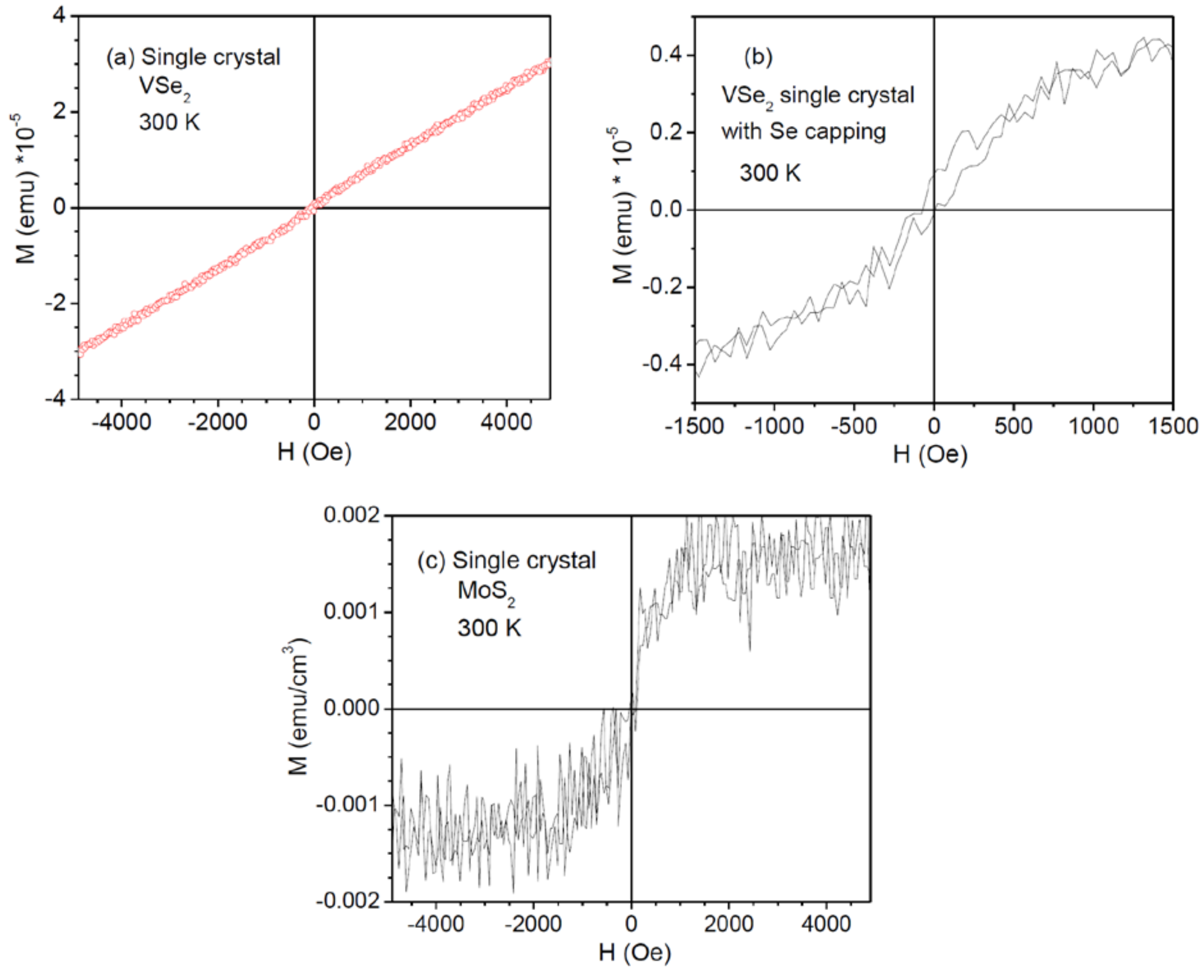

Fig. S4: Paramagnetism of bulk VSe_2 and weak magnetism of bulk MoS_2. (a) Linear field dependence of magnetization M(H) taken at 300 K indicates the paramagnetic nature of bulk VSe_2, which is in full agreement with the previous reports[1,2]. (b) The nearly linear behavior of the M-H curve taken at 300 K for bulk VSe_2 with a 10 nm Se capping layer excludes a surface effect as the origin of strong ferromagnetism. Therefore, the observation of intrinsic room temperature ferromagnetism in monolayers of VSe_2 grown on HOPG or MoS_2 substrates (Figs. 3 and 4b) indicates an important effect of reduced dimensionality on magnetic ordering in a TMD; (c) Bulk MoS_2 shows an extremely weak ferromagnetism, which is consistent with the previous observation[3]. This confirms that the room temperature ferromagnetism observed in monolayers of VSe_2 grown on MoS_2 substrates (Fig. 4) is intrinsic to the VSe_2 layer.

Supplementary Fig. 5

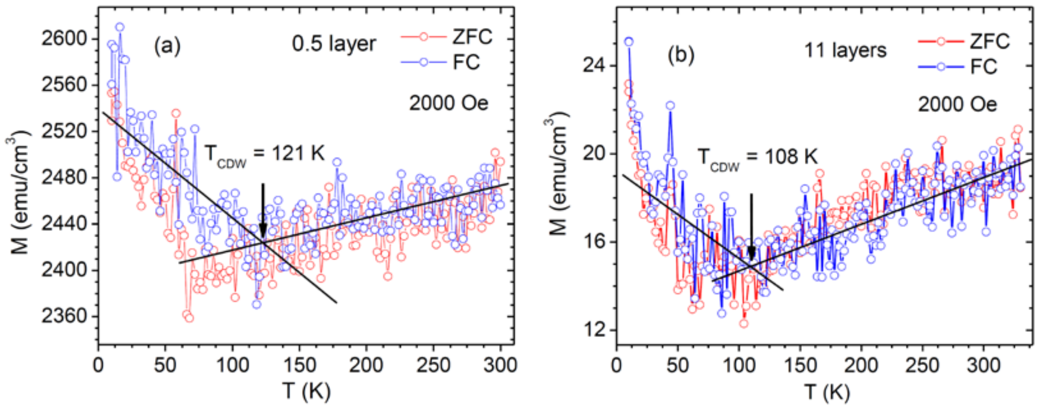

Fig. S5: Temperature dependence of zero-field-cooled (ZFC) and field-cooled (FC) magnetization of single- and multi-layers of VSe2: determination of CDW transition. Temperature dependences of zero-field-cooled (ZFC) and field-cooled (FC) magnetization of single- and multi-layers of VSe_2 show noted up-turns in the ZFC/FC magnetization with lowering temperature, which are defined as the charge density wave (CDW) transition temperatures T_{CDW}. T_{CDW} is 121 K and 108 K for 0.5 and 11 layers of VSe_2, respectively. These values of T_{CDW} match well with those determined from the temperature dependence of saturation magnetization (Fig. 3d).

References

[1]van Bruggen, C.F., Haas, C. Magnetic susceptibility and electrical properties of VSe_2 single crystals. *Solid State Commun.* **20**, 251-254 (1976).

[2] Bayard, M., Sienko, M.J. Anomalous electric and magnetic properties of vanadium diselenide. *J.Solid State Chem.* **19**, 325-329 (1976).

[3] Tongay, S., Varnoosfaderani, S.S., Appleton, B.R., Junqiao Wu, J.Q., Hebard, A.F., Magnetic properties of MoS2: Existence of ferromagnetism, *Appl. Phys. Lett.* **101**, 123105 (2012).