Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.
This journal is © the Owner Societies 2022

Supporting Information

Effects of Vacancy Defects on the Magnetic Properties of Vanadium Diselenide Monolayers: a First Principle Investigation

Yan Zhu, Yinlu Gao, Xue Jiang*, Jijun Zhao

Key Laboratory of Material Modification by Laser, Ion and Electron Beams, Dalian University of Technology, Ministry of Education, Dalian 116024, China

*Corresponding author. E-mail: jiangx@dlut.edu.cn (Xue Jiang)

Table S1. Vacancy formation energies (E_F) in V-rich/Se-rich condition, average magnetic moments (M), and V–Se bond lengths ($d_{V–Se}$) surrounding vacancies in the perfect and 45 kinds of defective VSe_2 monolayers (MLs).

Systems	E_F (eV)	M (μ_B/V)	$d_{V–Se}$ (Å)	Systems	E_F (eV)	M (μ_B/V)	$d_{V–Se}$ (Å)
VSe_2	---	0.675	2.486	D_{Se}-(9)	0.38/2.00	0.379	2.516
S_V	2.94/1.32	0.567	2.404	D_{Se}-(10)	0.11/1.73	0.183	2.514
S_{Se}	0.36/1.17	0.466	2.511	D_{Se}-(11)	0.86/2.48	0.315	2.516
D_V-(1)	5.11/1.87	0.390	2.431	D_{Se}-(12)	0.46/2.08	0.322	2.510
D_V-(2)	6.13/2.89	0.526	2.408	D_{Se}-(13)	0.46/2.08	0.670	2.459
D_V-(3)	6.13/2.87	0.529	2.412	D_{Se}-(14)	0.37/1.99	0.427	2.517
D_V-(4)	5.96/2.72	0.478	2.405	D_{Se}-(15)	0.67/2.29	0.707	2.490
D_{VSe}-(1)	2.35/1.54	0.461	2.421	T_{Se}-(1)(1)	1.34/3.77	0.677	2.434
D_{VSe}-(2)	2.87/2.06	0.370	2.449	T_{Se}-(2)(2)	1.17/3.60	0.681	2.434
D_{VSe}-(3)	2.99/2.18	0.440	2.441	T_{Se}-(6)(6)	0.82/3.25	0.279	2.514
D_{VSe}-(4)	3.00/2.19	0.481	2.648	T_{Se}-(10)(10)	0.75/3.18	0.213	2.504
D_{VSe}-(5)	3.18/2.37	0.476	2.439	T_{Se}-(11)(11)	1.74/4.17	0.800	2.519
D_{VSe}-(6)	3.13/2.32	0.228	2.452	T_{Se}-(1)(6)	0.95/3.38	0.700	2.437
D_{VSe}-(7)	3.34/2.53	0.416	2.284	T_{Se}-(10)(1)	0.36/2.79	0.502	2.472
D_{VSe}-(8)	3.17/2.36	0.292	2.455	T_{Se}-(3)(10)	1.34/3.77	0.404	2.516
D_{Se}-(1)	0.43/2.05	0.596	2.452	T_{Se}-(10)(2)	0.96/3.39	0.632	2.471
D_{Se}-(2)	0.66/2.28	0.751	2.426	Q_{Se}-(2)(4)(2)	2.42/5.66	0.723	2.455
D_{Se}-(3)	1.27/2.89	0.405	2.513	Q_{Se}-(2)(5)(2)	4.24/7.48	0.737	2.468
D_{Se}-(4)	0.45/2.07	0.270	2.517	Q_{Se}-(6)(2)(3)	3.11/6.35	0.803	2.460
D_{Se}-(5)	0.16/1.78	0.547	2.498	Q_{Se}-(2)(5)(9)	1.80/5.04	0.576	2.432
D_{Se}-(6)	0.76/2.38	0.411	2.499	Q_{Se}-(9)(6)(4)	1.50/4.74	0.322	2.521
D_{Se}-(7)	0.38/2.00	0.197	2.494	Q_{Se}-(17)(9)(17)	2.17/5.41	0.712	2.458
D_{Se}-(8)	0.32/1.94	0.534	2.494	Q_{Se}-(21)(10)(10)	0.97/4.21	0.208	2.507

Table S2. Electronic occupation numbers (N_{eo}) of d orbitals for the V atoms near the Se vacancies. The positive and negative values represent spin up and spin down, respectively.

	d_{yz}/d_{xz}	d_{yz}/d_{xz}	$d_{xy}/d_{x^2-y^2}$	$d_{xy}/d_{x^2-y^2}$	d_{z^2}	d_{z^2}
VSe_2	0.449	−0.345	0.364	−0.242	0.384	−0.172
S_{Se}	0.416	−0.384	0.338	−0.292	0.330	−0.279
D_{Se}-(10)	0.400	−0.400	0.294	−0.296	0.314	−0.311
T_{Se}-(10)(10)	0.404	−0.388	0.330	−0.321	0.333	−0.282
Q_{Se}-(21)(10)(10)	0.377	−0.376	0.326	−0.308	0.332	−0.291

Table S3. Electronic occupation numbers (N_{eo}) of d orbitals for the V atoms away from Se vacancies. The positive and negative values represent spin up and spin down, respectively.

	d_{yz}/d_{xz}	d_{yz}/d_{xz}	$d_{xy}/d_{x^2-y^2}$	$d_{xy}/d_{x^2-y^2}$	d_{z^2}	d_{z^2}
VSe_2	0.449	−0.345	0.364	−0.242	0.384	−0.172
S_{Se}	0.433	−0.359	0.347	−0.262	0.368	−0.210
D_{Se}-(10)	0.405	−0.374	0.330	−0.283	0.328	−0.277
T_{Se}-(10)(10)	0.427	−0.361	0.325	−0.272	0.348	−0.282
Q_{Se}-(21)(10)(10)	0.403	−0.371	0.326	−0.275	0.359	−0.286

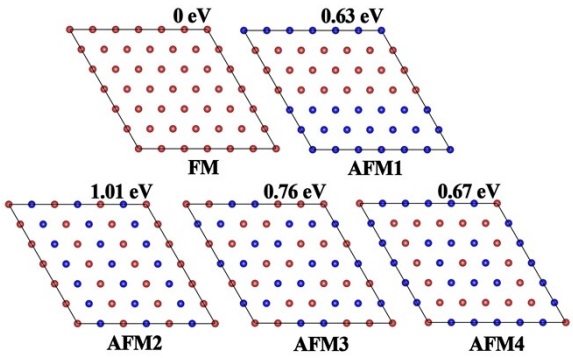

Figure S1. Spin arrangements of calculated ferromagnetic (FM) and antiferromagnetic (AFM) configurations. Red and blue balls represent V atoms in the states of spin up and spin down, respectively. The Se atoms are omitted for simplicity. The numbers represent the energy differences between the five configurations and the FM ordering.

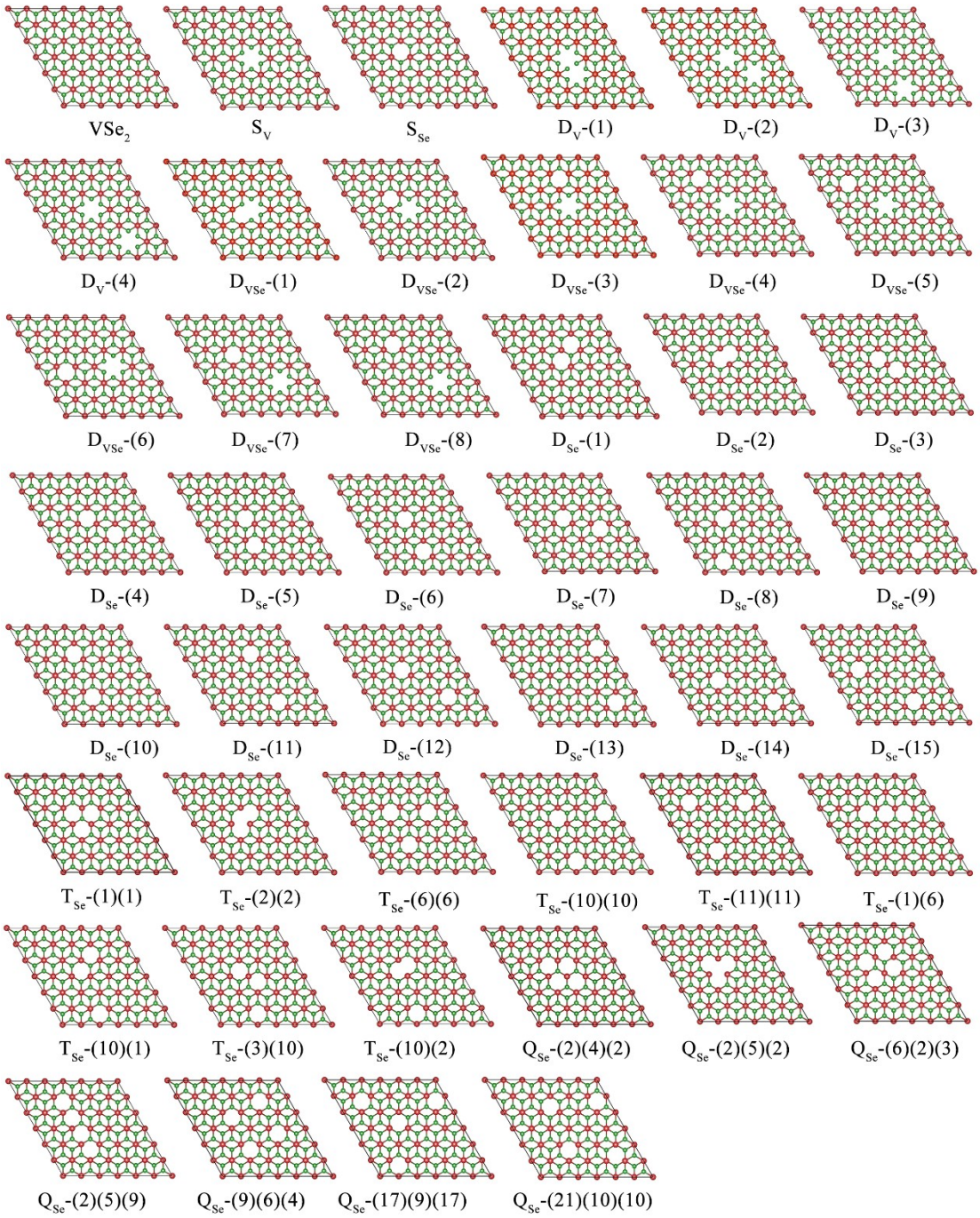

Figure S2. Geometric configurations of perfect VSe_2 monolayers and 45 kinds of vacancy structures.

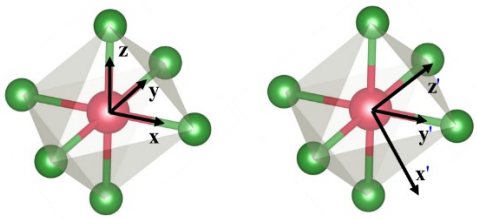

Figure S3. Conventional octahedral crystal structure with respect to the standard Cartesian coordinate x, y, z in 1T-VSe_2 (left); The same structure in the rotated Cartesian coordinate system x', y', z' in triangular field (right).