Single-atom vanadium-doped 2D semiconductor platform for attomolar-level molecular sensing

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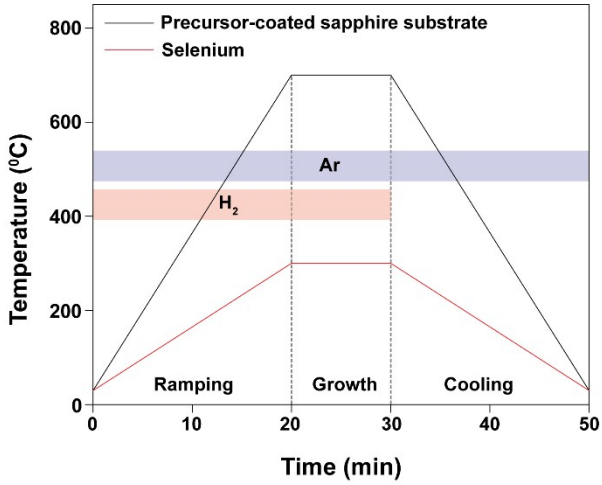

Fig. S1 Temperature profiles and gas composition in the liquid precursor-based CVD of pristine $ReSe_2$ and $V_{SAD}ReSe_2$.

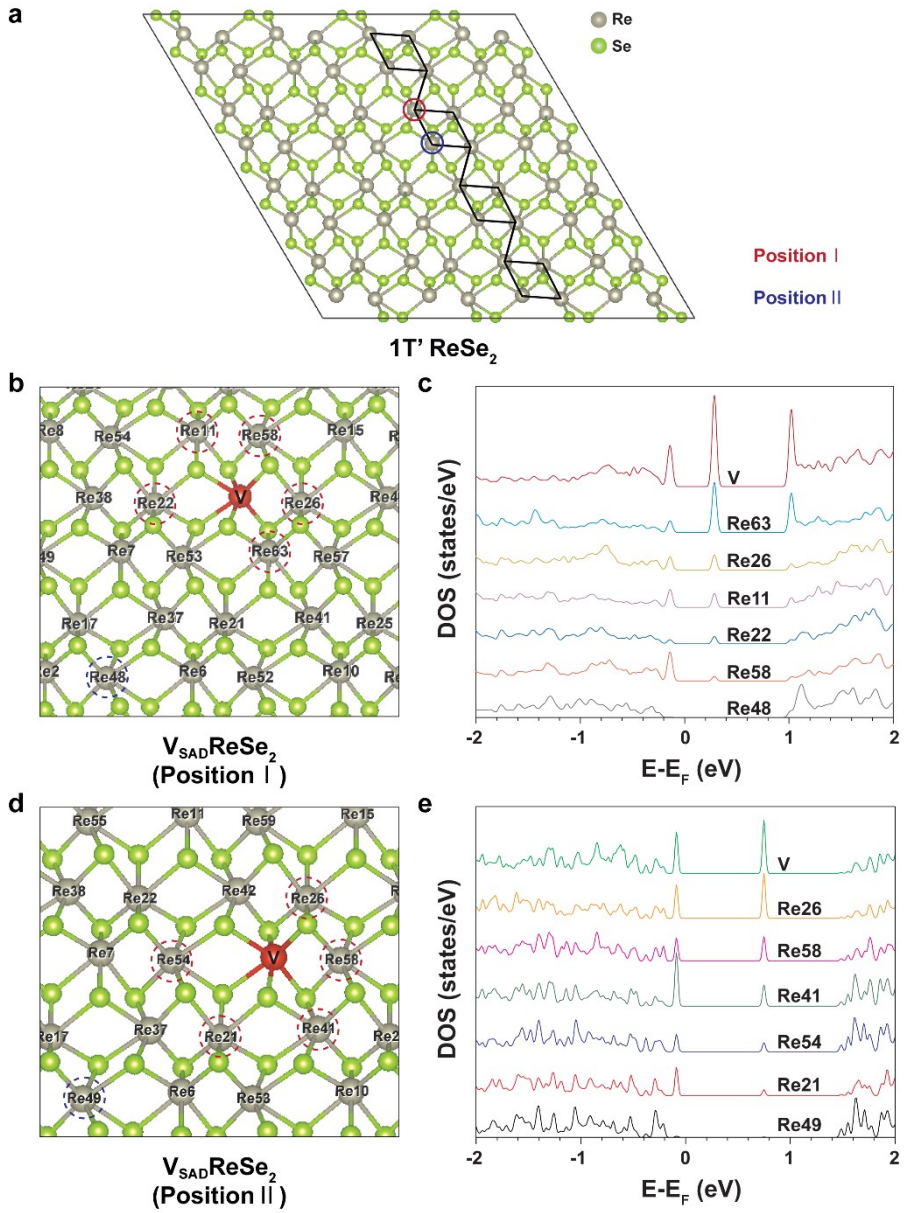

Fig. S2 (a) Two position types for the substitutional doping of vanadium atoms in the $V_{SAD}ReSe_2$ lattice. (b) Atomic structure of $V_{SAD}ReSe_2$ for position I. (c) Projected DOS of the vanadium atom and neighboring rhenium atoms in $V_{SAD}ReSe_2$ (position I). (d) Atomic structure of $V_{SAD}ReSe_2$ for position II. (e) Projected DOS of the vanadium atom and neighboring rhenium atoms in $V_{SAD}ReSe_2$ (position II).

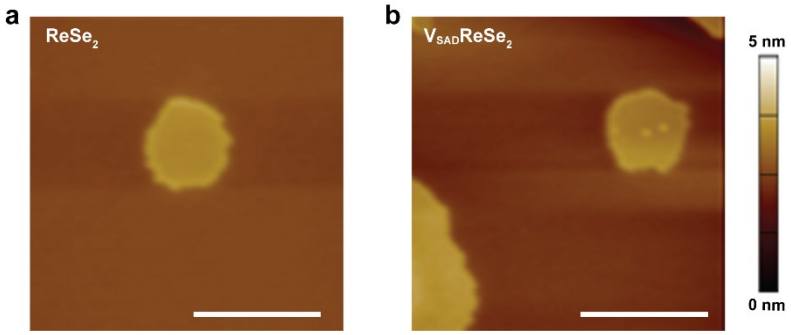

Fig. S3 AFM images of pristine $ReSe_2$ and $V_{SAD}ReSe_2$ synthesized by liquid precursor-assisted CVD (scale bars: 4 μm).

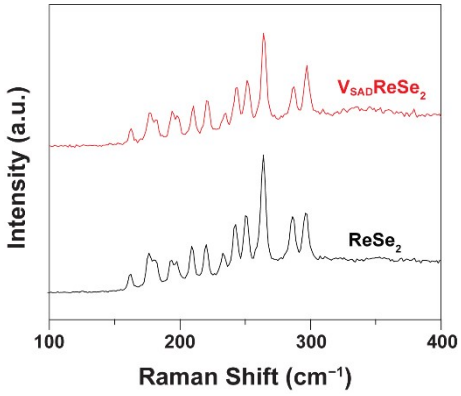

Fig. S4 Raman spectra of pristine $ReSe_2$ and $V_{SAD}ReSe_2$ synthesized by liquid precursor-assisted CVD.

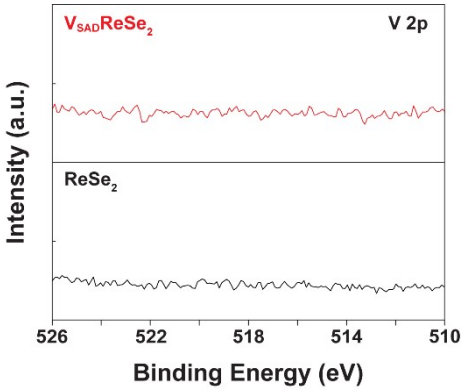

Fig. S5 XPS spectra of V 2p obtained from pristine $ReSe_2$ and $V_{SAD}ReSe_2$.

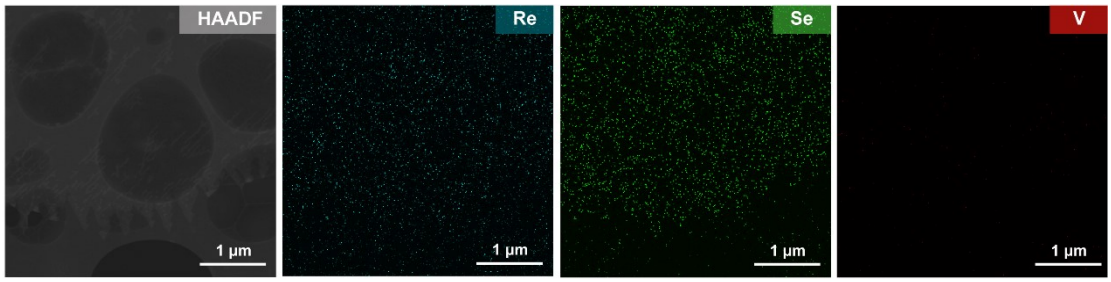

Fig. S6 EDS mapping image of pristine $ReSe_2$ for Re, Se, and V.

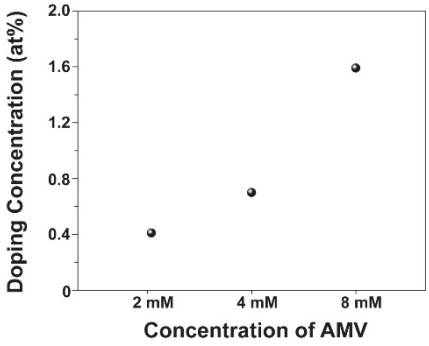

Fig. S7 Doping concentration of $V_{SAD}ReSe_2$ synthesized with different vanadium precursor concentrations.

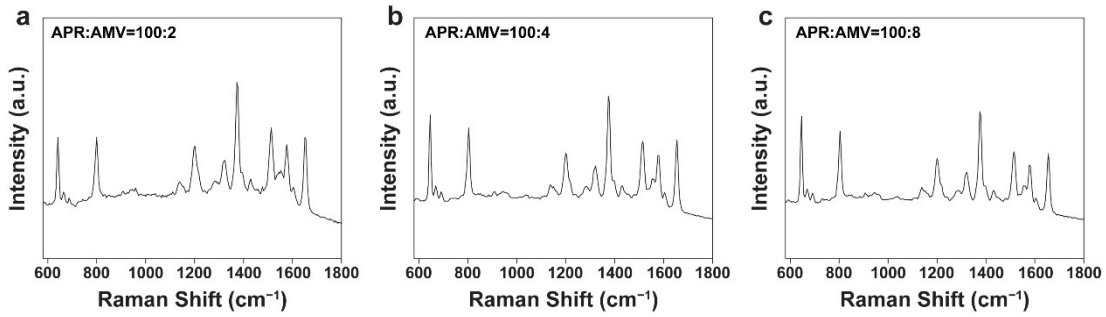

Fig. S8 SERS profiles for 10^{-4} M of R6G on the as-synthesized $V_{SAD}ReSe_2$ with different precursor ratios (APR:AMV=100 mM : X mM, X = 2, 4, and, 8).

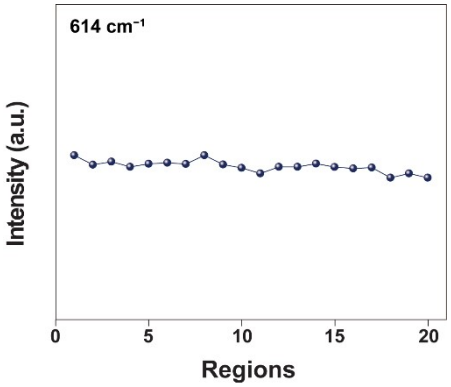

Fig. S9 Raman-signal intensity of R6G molecules on $V_{SAD}ReSe_2$ at 614 cm^{-1} for 20 different regions in the samples shown in Fig. 4d.

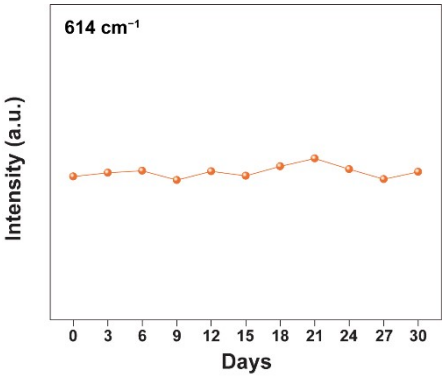

Fig. S10 Raman-signal intensity of R6G molecules on $V_{SAD}ReSe_2$ at 614 cm^{-1} measured for up to 30 days.

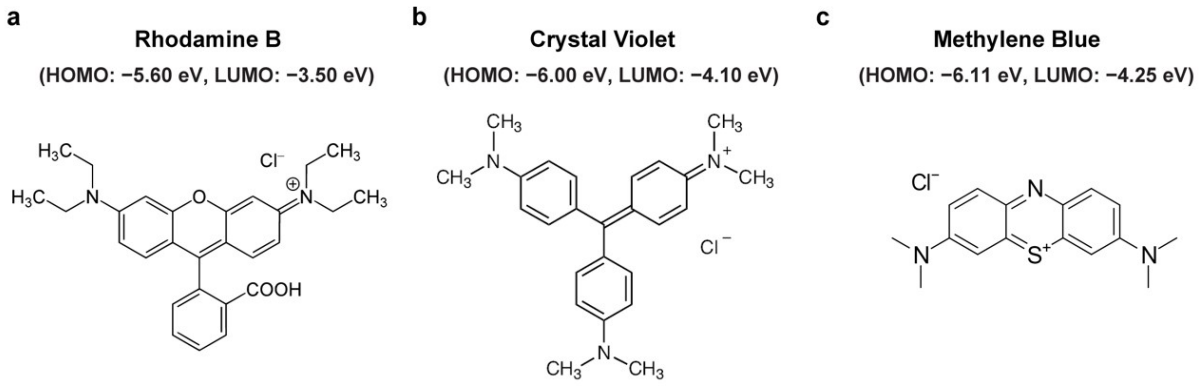

Fig. S11 HOMO-LUMO energy levels and molecular structures of (a) rhodamine B, (b) crystal violet, and (c) methylene blue.

Table S1 Raman peaks and peak assignments of the R6G molecule.

Raman peak	Peak assignment
614 cm^{-1}	C-C-C ring in-plane bending
776 cm^{-1}	C-H out-of-plane bending
1131 cm^{-1}	C-H in-plane bending
1185 cm^{-1}	C-C stretching vibration bending
1312 cm^{-1}	Aromatic C-C stretching
1363 cm^{-1}	Aromatic C-C stretching
1419 cm^{-1}	Aromatic C-C stretching / C-H vibration
1506 cm^{-1}	Aromatic C-C stretching
1532 cm^{-1}	Aromatic C-C stretching
1575 cm^{-1}	Aromatic C-C stretching
1601 cm^{-1}	Aromatic C-C stretching / C-H vibration
1650 cm^{-1}	Aromatic C-C stretching

Table S2 Summary of the SERS performance for 2D material-based SERS substrates reported in literature.

SERS substrate	Synthesis method	Probe molecule	LOD	Excitation wavelength	Reference
Oxygen-substituted MoS_2	Hydrothermal synthesis	R6G	1×10^{-7} M	532 nm	1
Mildly reduced GO	Modified Hummers' method	RhB	5×10^{-8} M	514 nm	2
1T-$MoSe_2$ (*n*-butyl lithium)	Chemical exfoliation	R6G	1×10^{-8} M	532 nm	3
1T-MoS_2 (NaK)	Chemical exfoliation	CV	1×10^{-8} M	532 nm	4
ReS_2	CVD	R6G, MB	1×10^{-9} M	532, 633 nm	5
MoS_2 QD/rGO	Solvothermal method/Modified Hummers' method	R6G	1×10^{-9} M	532 nm	6
AuNPs/MoS_2	CVD	RhB	1×10^{-10} M	532 nm	7
N-doped graphene	CVD	RhB	1×10^{-11} M	514 nm	8
1T'-W(Mo)Te_2	CVD	R6G	$4(40) \times 10^{-14}$ M	532 nm	9
NbS_2	CVD	MeB	1×10^{-14} M	532 nm	10
Graphene/ReO_xS_y	CVD	R6G	1×10^{-15} M	532 nm	11
Ti_2N	Selective etching of Al from Ti_2AlN (MAX)	R6G	1×10^{-15} M	532 nm	12
$V_{SAD}ReSe_2$	**CVD**	**R6G**	**1×10^{-18} M**	**532 nm**	**This work**

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