

Supporting Information

Direct Detection of Inhomogeneity in CVD-Grown 2D TMD Materials via K-Means Clustering Raman Analysis

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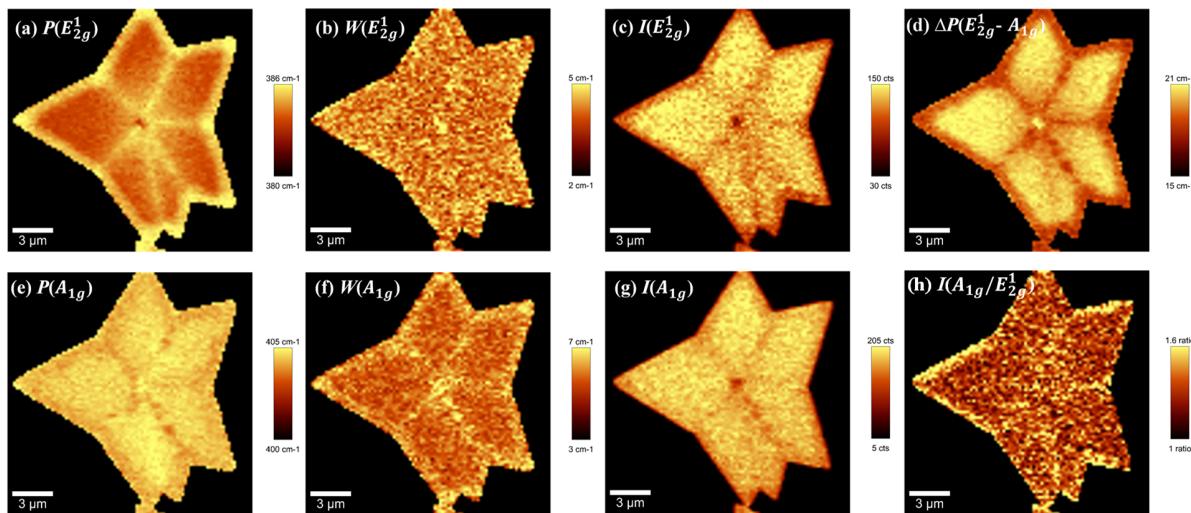


Figure S1. (a-c) and (e-g) are Raman images of monolayer polycrystalline MoS₂ plotted as a function of the peak position (P), width (W), and intensity (I) of E_{2g}^1 and A_{1g} modes. The difference between E_{2g}^1 and A_{1g} modes in peak position and relative peak intensities are calculated and plotted in (d) and (h).

Cluster Name	$P(E_{2g}^1)$ (cm $^{-1}$)	$W(E_{2g}^1)$ (cm $^{-1}$)	$I(E_{2g}^1)$ (a.u.)	$P(A_{1g})$ (cm $^{-1}$)	$W(A_{1g})$ (cm $^{-1}$)	$I(A_{1g})$ (a.u.)	$\Delta P(A_{1g}-E_{2g}^1)$ (cm $^{-1}$)	$I(A_{1g}/E_{2g}^1)$
Cluster 1	387.96	4.55	108.50	406.99	6.26	154.86	19.03	1.43
Cluster 2	387.29	4.39	127.91	407.25	5.92	177.87	19.96	1.39
Cluster 3	386.63	4.34	140.65	407.25	5.70	185.79	20.61	1.32

Table S1. Raman parameters of the cluster 1-3 in monolayer polycrystalline MoS₂ sample fitted with Lorentz function.

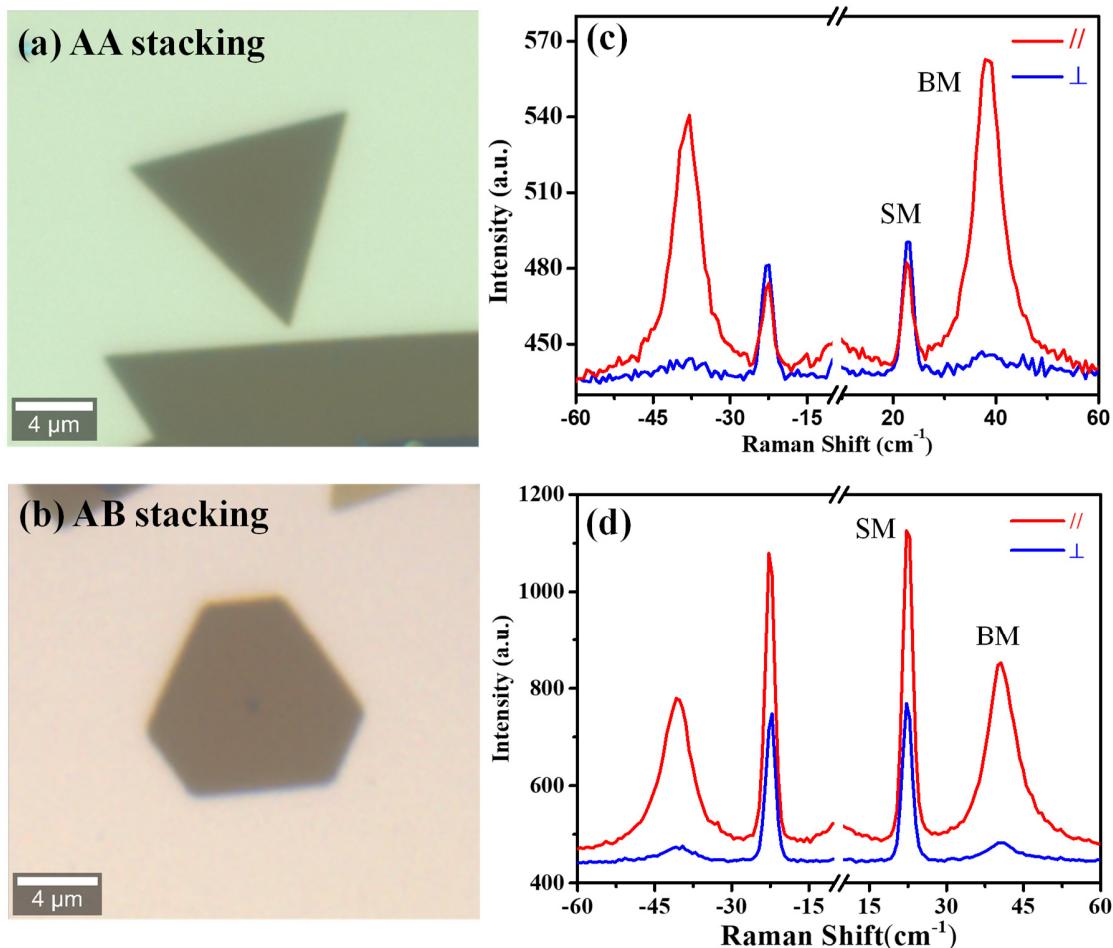


Figure S2. Stacking identification of AA and AB bilayer MoS₂ by polarized Raman spectroscopy. (a-b) Optical image of CVD-grown bilayer MoS₂ and (c-d) the correlated Stoke and anti-Stoke low wavenumber Raman spectra measured under parallel (//) and cross (⊥) polarization configurations, respectively. The AA and AB stacking orders could be extracted from the relative intensities between the shearing mode (SM) and breathing mode (BM) at 22.5 and 40.5 cm $^{-1}$ in excellent agreement with X. Yan *et al*'s work.¹

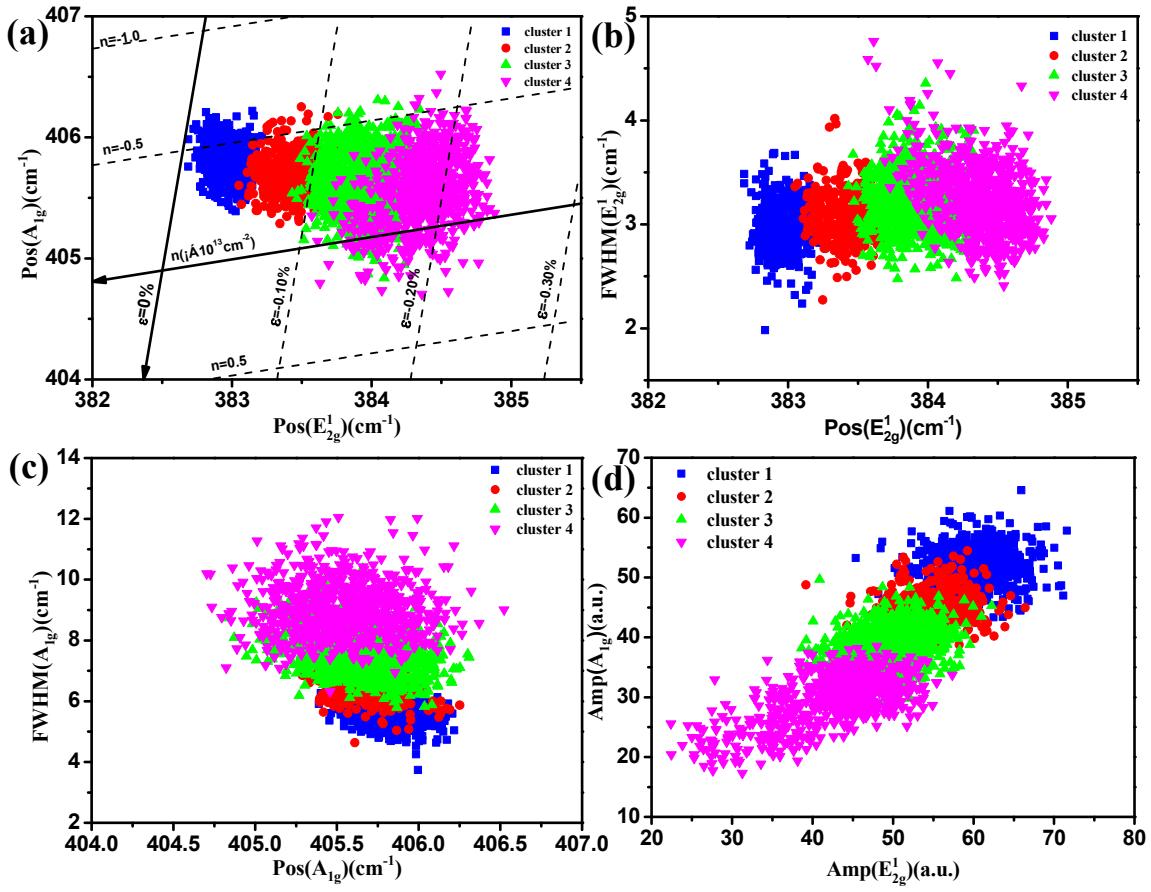


Figure S3. Correlative analysis of Raman spectral features to quantitatively evaluate the contribution of strain and doping effects on bilayer MoS₂ with AA-stacking. **(a)** The 2D correlative analysis of $\text{Pos}(E_{2g}^1)$ and $\text{Pos}(A_{1g})$ in cluster 1-4. **(b)** Correlation plot of $\text{FWHM}(E_{2g}^1)$ vs $\text{Pos}(E_{2g}^1)$. **(c)** Correlation plot of $\text{FWHM}(A_{1g})$ vs $\text{Pos}(A_{1g})$. **(d)** Correlation plot of $\text{AMP}(A_{1g})$ vs $\text{AMP}(E_{2g}^1)$.

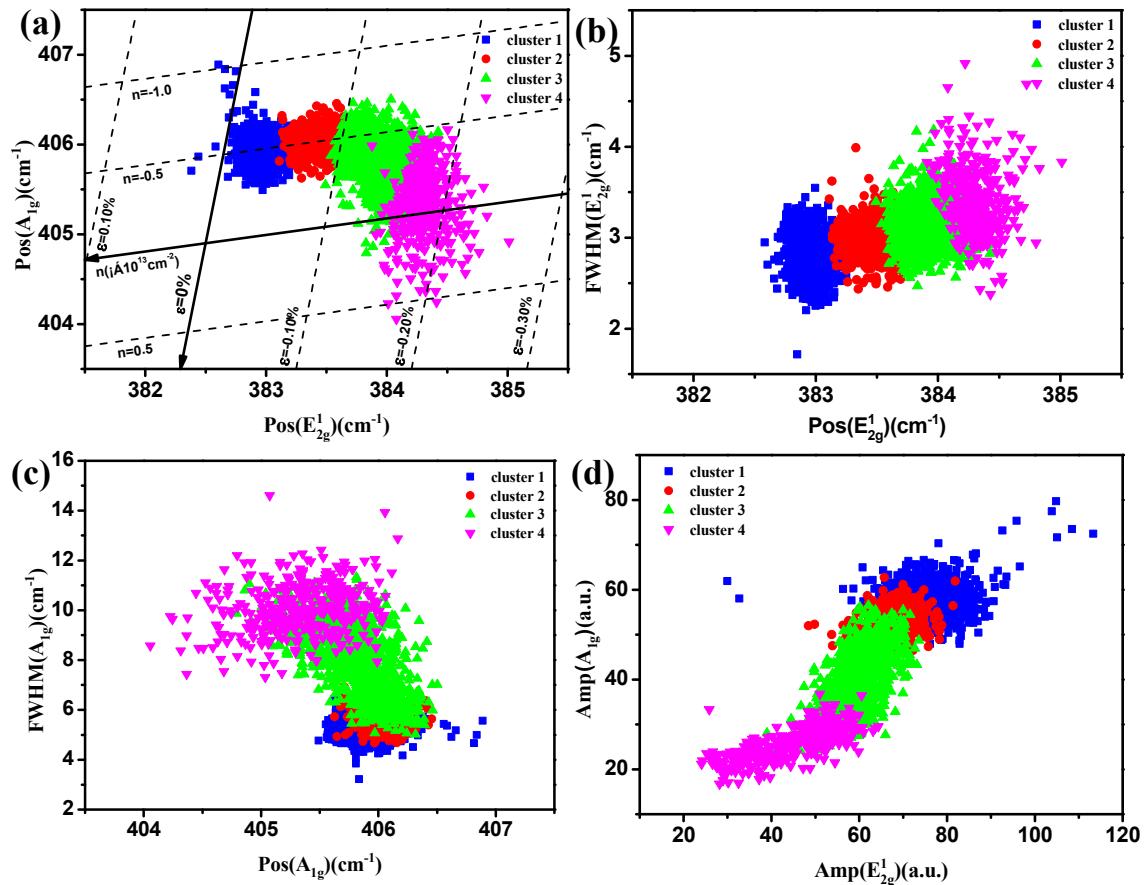


Figure S4. Correlative analysis of Raman spectral features to quantitatively evaluate the contribution of strain and doping effects on monolayer MoS₂ with AB stacking. The 2D correlative analysis of (a) Correlation plots of $\text{Pos}(E_{2g}^1)$ and $\text{Pos}(A_{1g})$ in cluster 1-4. (b)~(d) Correlation plots of $\text{FWHM}(E_{2g}^1)$ vs $\text{Pos}(E_{2g}^1)$, $\text{FWHM}(A_{1g})$ vs $\text{Pos}(A_{1g})$ and $\text{AMP}(A_{1g})$ vs $\text{AMP}(E_{2g}^1)$, respectively.

1 Yan, J. *et al.* Stacking-Dependent Interlayer Coupling in Trilayer MoS(2) with Broken Inversion Symmetry. *Nano Lett* **15**, 8155-8161, doi:10.1021/acs.nanolett.5b03597 (2015).