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Supplementary Information

Interlayer bond polarizability model for stacking-dependent low-frequency Raman scattering in layered materials

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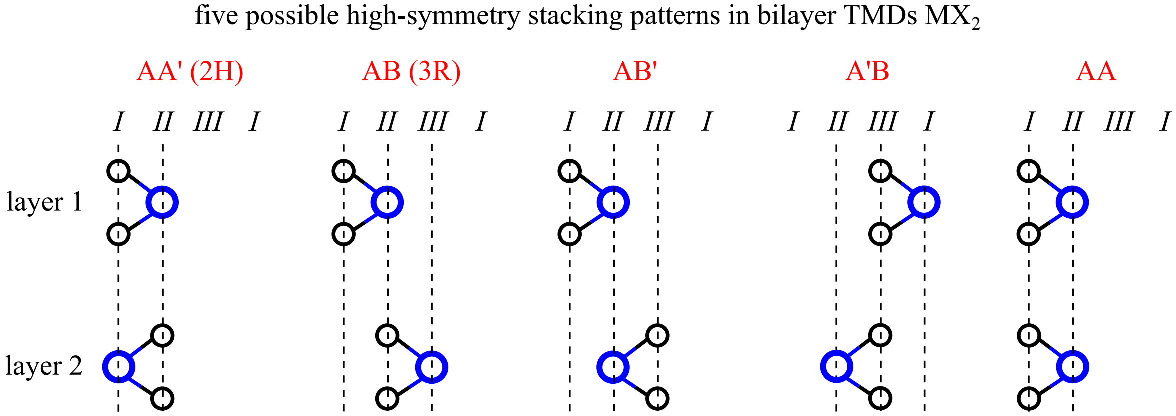

Figure S1 Side views of the five possible high-symmetry stacking patterns in bilayer TMDs MX$_2$ like MoS$_2$. The blue (black) circles represent metal M (chalcogenide X) atoms. In the honeycomb lattice, there are three atomic coordinates: *I*: 0, 0; *II*: 1/3, 2/3; *III*: 2/3, 1/3. In bilayer MX$_2$, AA$'$ (corresponding to bulk 2H stacking) and AB (corresponding to bulk 3R stacking) are stable and can be commonly found in natural and synthetic samples.

S1 Generalized interlayer bond polarizability model

In this section, we present the detailed derivation process of the generalized bond polarizability model.[1–4] The Raman intensity of a phonon mode k is given by[1,4,5]

$$I(k) \propto \left|\boldsymbol{e}_{\mathrm{i}} \cdot \tilde{\boldsymbol{R}}(k) \cdot \boldsymbol{e}_{\mathrm{s}}^{T}\right|^{2} \propto \left|\sum_{\mu\nu} e_{\mathrm{i},\mu} e_{\mathrm{s},\nu} \Delta\alpha_{\mu\nu}(k)\right|^{2}, \tag{S1}$$

where $\tilde{\boldsymbol{R}}(k)$ is the (3×3) Raman tensor of the phonon mode k, subscripts μ and ν indicate Cartesian components (x, y or z) of the tensor, and $\boldsymbol{e}_{\mathrm{i}}$ and $\boldsymbol{e}_{\mathrm{s}}$ are the unit vectors for the polarization of the incident and scattered light, respectively. The Raman tensor element

$$\Delta\alpha_{\mu\nu}(k) = \sum_{j\gamma} \left[\frac{\partial \alpha_{\mu\nu}}{\partial r_{j\gamma}}\right]_0 \Delta r_{j\gamma}(k), \tag{S2}$$

where $r_{j\gamma}$ is the position of atom j along direction γ (x, y or z) in equilibrium, $\left[\frac{\partial \alpha_{\mu\nu}}{\partial r_{j\gamma}}\right]_0$ is the derivative of the electronic polarizability tensor element $\alpha_{\mu\nu}$ with respect to the atomic displacement from the equilibrium configuration, and $\Delta r_{j\gamma}(k)$ is the eigen-displacement of atom j along direction γ in the phonon mode k (i.e., the eigenvector of the mass-normalized dynamic matrix).[5] One can see that the Raman tensor of the phonon mode k is proportional to the change of the polarizability by its vibration. According to the empirical bond polarizability model, the polarizability of the system can be approximated by a sum of individual bond polarizabilities from different bonds:[1,4]

$$\alpha_{\mu\nu} = \frac{1}{2}\sum_{iB}\left[\frac{\alpha_{\parallel,B}+2\alpha_{\perp,B}}{3}\delta_{\mu\nu} + (\alpha_{\parallel,B}-\alpha_{\perp,B})\left(\frac{R_{i\mu,B}R_{i\nu,B}}{R_{i,B}^{2}} - \frac{1}{3}\delta_{\mu\nu}\right)\right], \tag{S3}$$

where B indicates a bond connected to atom i, the boldface $\boldsymbol{R}_{i,B}$ is the corresponding bond vector connecting atom i to one of its neighbor atoms i', $R_{i\mu,B}$ is the μ (x, y or z) component of $\boldsymbol{R}_{i,B}$, and $R_{i,B}$ is the length of $\boldsymbol{R}_{i,B}$. $\alpha_{\parallel,B}$ and $\alpha_{\perp,B}$ are the bond polarizabilities for the bond B in the directions parallel and perpendicular to the bond, respectively. They depend on the bond $R_{i,B}$ and therefore,

$$\begin{aligned} \frac{\partial \alpha_{\parallel,B}}{\partial r_{j\gamma}} &= \frac{\partial \alpha_{\parallel,B}}{\partial R_{i,B}}\frac{\partial R_{i,B}}{\partial r_{j\gamma}} = \alpha'_{\parallel,B}\frac{\partial R_{i,B}}{\partial r_{j\gamma}} \\ \frac{\partial \alpha_{\perp,B}}{\partial r_{j\gamma}} &= \frac{\partial \alpha_{\perp,B}}{\partial R_{i,B}}\frac{\partial R_{i,B}}{\partial r_{j\gamma}} = \alpha'_{\perp,B}\frac{\partial R_{i,B}}{\partial r_{j\gamma}}, \end{aligned} \tag{S4}$$

where $\alpha'_{\parallel,B}$ and $\alpha'_{\perp,B}$ are the radial derivatives of the bond polarizabilities with respect to the bond length. The values of $\alpha_{\parallel,B}$, $\alpha_{\perp,B}$, $\alpha'_{\parallel,B}$ and $\alpha'_{\perp,B}$ are functions of the bond length, and usually determined by fitting with experimental Raman intensities.[1–4]

To obtain $\frac{\partial R_{i,B}}{\partial r_{j\gamma}}$, we need to use the following relations: $\boldsymbol{R}_{i,B} = \vec{r}_{i'} - \vec{r}_i, \boldsymbol{R}_{i',B} = \vec{r}_i - \vec{r}_{i'} = -\boldsymbol{R}_{i,B}, R_{i,B} = \sqrt{\sum_\eta (r_{i'\eta} - r_{i\eta})^2} = R_{i',B}$. Subsequently,

$$\begin{aligned} &\sum_{j\gamma} \frac{\partial R_{i,B}}{\partial r_{j\gamma}} \Delta r_{j\gamma}(k) = \sum_{j\gamma} \frac{\partial \sqrt{\sum_\eta (r_{i'\eta} - r_{i\eta})^2}}{\partial r_{j\gamma}} \Delta r_{j\gamma}(k) = \sum_{j\gamma} \frac{1}{2} \frac{1}{\sqrt{\sum_\eta (r_{i'\eta} - r_{i\eta})^2}} \frac{\partial \sum_\eta (r_{i'\eta} - r_{i\eta})^2}{\partial r_{j\gamma}} \Delta r_{j\gamma}(k) \\ &= \sum_{j\gamma} \frac{1}{2} \frac{1}{\sqrt{\sum_\eta (r_{i'\eta} - r_{i\eta})^2}} \left(\sum_\eta 2(r_{i'\eta} - r_{i\eta}) \frac{\partial (r_{i'\eta} - r_{i\eta})}{\partial r_{j\gamma}} \right) \Delta r_{j\gamma}(k) \qquad (\eta \text{ or } \gamma = x, y, z) \\ &= \sum_{j\gamma} \frac{1}{2} \frac{1}{\sqrt{\sum_\eta (r_{i'\eta} - r_{i\eta})^2}} \left(\sum_\eta 2(r_{i'\eta} - r_{i\eta})(\delta_{i'j}\delta_{\eta\gamma} - \delta_{ij}\delta_{\eta\gamma}) \right) \Delta r_{j\gamma}(k) \\ &= \sum_{j\gamma} \frac{1}{R_{i,B}} \left[(r_{i'\gamma} - r_{i\gamma})(\delta_{i'j} - \delta_{ij}) \right] \Delta r_{j\gamma}(k) = \sum_{j\gamma} \frac{(r_{i'\gamma} - r_{i\gamma})\delta_{i'j}}{R_{i,B}} \Delta r_{j\gamma}(k) - \sum_{j\gamma} \frac{(r_{i'\gamma} - r_{i\gamma})\delta_{ij}}{R_{i,B}} \Delta r_{j\gamma}(k) \\ &= \sum_\gamma \frac{(r_{i'\gamma} - r_{i\gamma})}{R_{i,B}} \Delta r_{i'\gamma}(k) - \sum_\gamma \frac{(r_{i'\gamma} - r_{i\gamma})}{R_{i,B}} \Delta r_{i\gamma}(k) = -\sum_\gamma \frac{(r_{i\gamma} - r_{i'\gamma})}{R_{i,B}} \Delta r_{i'\gamma}(k) - \sum_\gamma \frac{(r_{i'\gamma} - r_{i\gamma})}{R_{i,B}} \Delta r_{i\gamma}(k) \\ &= -\frac{(\vec{r}_i - \vec{r}_{i'})}{R_{i,B}} \cdot \Delta\vec{r}_{i'}(k) - \frac{(\vec{r}_{i'} - \vec{r}_i)}{R_{i,B}} \cdot \Delta\vec{r}_i(k) = -\frac{\boldsymbol{R}_{i',B}}{R_{i',B}} \cdot \Delta\vec{r}_{i'}(k) - \frac{\boldsymbol{R}_{i,B}}{R_{i,B}} \cdot \Delta\vec{r}_i(k) \qquad (R_{i',B} = R_{i,B}) \\ &\qquad\qquad \Downarrow \\ &\sum_{iB}\sum_{j\gamma} \frac{\partial R_{i,B}}{\partial r_{j\gamma}} \Delta r_{j\gamma}(k) = -\sum_{iB} \left(\frac{\boldsymbol{R}_{i',B}}{R_{i',B}} \cdot \Delta\vec{r}_{i'}(k) \right) - \sum_{iB} \left(\frac{\boldsymbol{R}_{i,B}}{R_{i,B}} \cdot \Delta\vec{r}_i(k) \right) = -2\sum_{iB} \left(\frac{\boldsymbol{R}_{i,B}}{R_{i,B}} \cdot \Delta\vec{r}_i(k) \right), \end{aligned} \tag{S5}$$

where $\sum_{iB}\left(\frac{\boldsymbol{R}_{i',B}}{R_{i',B}}\cdot\Delta\vec{r}_{i'}(k)\right)=\sum_{ii'B}\left(\frac{\boldsymbol{R}_{i',B}}{R_{i',B}}\cdot\Delta\vec{r}_{i'}(k)\right)=\sum_{i'B}\left(\frac{\boldsymbol{R}_{i',B}}{R_{i',B}}\cdot\Delta\vec{r}_{i'}(k)\right)=\sum_{iB}\left(\frac{\boldsymbol{R}_{i,B}}{R_{i,B}}\cdot\Delta\vec{r}_{i}(k)\right)$. Furthermore,

$$\begin{aligned}
\sum_{iB}\sum_{j\gamma}\frac{\partial}{\partial r_{j\gamma}}\left(\frac{1}{R_{i,B}^2}\right)\Delta r_{j\gamma}(k) &= \sum_{iB}\sum_{j\gamma}\left(-\frac{2}{R_{i,B}^3}\frac{\partial R_{i,B}}{\partial r_{j\gamma}}\Delta r_{j\gamma}(k)\right)\\
&= \sum_{iB}-\frac{2}{R_{i,B}^3}\left(\sum_{j\gamma}\frac{\partial R_{i,B}}{\partial r_{j\gamma}}\Delta r_{j\gamma}(k)\right)\\
&= \sum_{iB}-\frac{2}{R_{i,B}^3}\left(-\frac{\boldsymbol{R}_{i',B}}{R_{i',B}}\cdot\Delta\vec{r}_{i'}(k)-\frac{\boldsymbol{R}_{i,B}}{R_{i,B}}\cdot\Delta\vec{r}_{i}(k)\right)\text{(see Eq. S5)}\\
&= 2\sum_{iB}\left(\frac{\boldsymbol{R}_{i',B}}{R_{i',B}^4}\cdot\Delta\vec{r}_{i'}(k)+\frac{\boldsymbol{R}_{i,B}}{R_{i,B}^4}\cdot\Delta\vec{r}_{i}(k)\right)\quad(R_{i',B}=R_{i,B})\\
&= 4\sum_{iB}\frac{\boldsymbol{R}_{i,B}}{R_{i,B}^4}\cdot\Delta\vec{r}_{i}(k),
\end{aligned}\tag{S6}$$

where similarly $\sum_{iB}\frac{\boldsymbol{R}_{i',B}}{R_{i',B}^4}\cdot\Delta\vec{r}_{i'}(k)=\sum_{iB}\frac{\boldsymbol{R}_{i,B}}{R_{i,B}^4}\cdot\Delta\vec{r}_{i}(k)$. In addition, the μ (x, y or z) component of $\boldsymbol{R}_{i,B}$ is $R_{i\mu,B}=r_{i'\mu}-r_{i\mu}$, and similarly $R_{i\nu,B}=r_{i'\nu}-r_{i\nu}$. It follows that

$$\begin{aligned}
&\sum_{j\gamma}\frac{\partial(R_{i\mu,B}R_{i\nu,B})}{\partial r_{j\gamma}}\Delta r_{j\gamma}(k)=\sum_{j\gamma}\frac{\partial(r_{i'\mu}-r_{i\mu})}{\partial r_{j\gamma}}R_{i\nu,B}\Delta r_{j\gamma}(k)+\sum_{j\gamma}R_{i\mu,B}\frac{\partial(r_{i'\nu}-r_{i\nu})}{\partial r_{j\gamma}}\Delta r_{j\gamma}(k)\\
&=\sum_{j\gamma}(\delta_{i'j}\delta_{\mu\gamma}-\delta_{ij}\delta_{\mu\gamma})R_{i\nu,B}\Delta r_{j\gamma}(k)+\sum_{j\gamma}R_{i\mu,B}(\delta_{i'j}\delta_{\nu\gamma}-\delta_{ij}\delta_{\nu\gamma})\Delta r_{j\gamma}(k)\\
&=\sum_{j}(\delta_{i'j}-\delta_{ij})R_{i\nu,B}\Delta r_{j\mu}(k)+\sum_{j}R_{i\mu,B}(\delta_{i'j}-\delta_{ij})\Delta r_{j\nu}(k)\\
&=\left(R_{i\nu,B}\Delta r_{i'\mu}(k)-R_{i\nu,B}\Delta r_{i\mu}(k)\right)+\left(R_{i\mu,B}\Delta r_{i'\nu}(k)-R_{i\mu,B}\Delta r_{i\nu}(k)\right)\\
&=\left(-R_{i'\nu,B}\Delta r_{i'\mu}(k)-R_{i\nu,B}\Delta r_{i\mu}(k)\right)+\left(-R_{i'\mu,B}\Delta r_{i'\nu}(k)-R_{i\mu,B}\Delta r_{i\nu}(k)\right)\qquad(R_{i\nu,B}=-R_{i'\nu,B};R_{i\mu,B}=-R_{i'\mu,B})\\
&\qquad\qquad\Downarrow\\
&\sum_{iB}\sum_{j\gamma}\frac{\partial(R_{i\mu,B}R_{i\nu,B})}{\partial r_{j\gamma}}\Delta r_{j\gamma}(k)=\sum_{iB}\left(-R_{i'\nu,B}\Delta r_{i'\mu}(k)-R_{i\nu,B}\Delta r_{i\mu}(k)\right)+\sum_{iB}\left(-R_{i'\mu,B}\Delta r_{i'\nu}(k)-R_{i\mu,B}\Delta r_{i\nu}(k)\right)\\
&=-2\sum_{iB}\left(R_{i\nu,B}\Delta r_{i\mu}(k)+R_{i\mu,B}\Delta r_{i\nu}(k)\right),
\end{aligned}\tag{S7}$$

where similarly, $\sum_{iB}R_{i'\nu,B}\Delta r_{i'\mu}(k)=\sum_{ii'B}R_{i'\nu,B}\Delta r_{i'\mu}(k)=\sum_{i'B}R_{i'\nu,B}\Delta r_{i'\mu}(k)=\sum_{iB}R_{i\nu,B}\Delta r_{i\mu}(k)$, and $\sum_{iB}R_{i'\mu,B}\Delta r_{i'\nu}(k)=\sum_{iB}R_{i\mu,B}\Delta r_{i\nu}(k)$.

With Eqs. S4, S5, S6 and S7, we then substitute Eq. S3 into Eq. S2, which yields the Raman tensor element

$$\begin{aligned}
\Delta\alpha_{\mu\nu}(k) &= \sum_{j\gamma}\frac{\partial}{\partial r_{j\gamma}}\left\{\frac{1}{2}\sum_{iB}\left[\frac{\alpha_{\parallel,B}+2\alpha_{\perp,B}}{3}\delta_{\mu\nu}+(\alpha_{\parallel,B}-\alpha_{\perp,B})\left(\frac{R_{i\mu,B}R_{i\nu,B}}{R_{i,B}^2}-\frac{1}{3}\delta_{\mu\nu}\right)\right]\right\}\Delta r_{j\gamma}(k)\\
&=\frac{1}{2}\sum_{iB}\left\{\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu}+(\alpha'_{\parallel,B}-\alpha'_{\perp,B})\left(\frac{R_{i\mu,B}R_{i\nu,B}}{R_{i,B}^2}-\frac{1}{3}\delta_{\mu\nu}\right)\right]\sum_{j\gamma}\frac{\partial R_{i,B}}{\partial r_{j\gamma}}\Delta r_{j\gamma}(k)\right\}\\
&\quad+\frac{1}{2}\sum_{iB}\left\{(\alpha_{\parallel,B}-\alpha_{\perp,B})\sum_{j\gamma}\frac{\partial}{\partial r_{j\gamma}}\left(\frac{R_{i\mu,B}R_{i\nu,B}}{R_{i,B}^2}\right)\Delta r_{j\gamma}(k)\right\}\\
&=\frac{1}{2}\sum_{iB}\left\{\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu}+(\alpha'_{\parallel,B}-\alpha'_{\perp,B})\left(\frac{R_{i\mu,B}R_{i\nu,B}}{R_{i,B}^2}-\frac{1}{3}\delta_{\mu\nu}\right)\right]\sum_{j\gamma}\frac{\partial R_{i,B}}{\partial r_{j\gamma}}\Delta r_{j\gamma}(k)\right\}\\
&\quad+\frac{1}{2}\sum_{iB}\left\{(\alpha_{\parallel,B}-\alpha_{\perp,B})\left[\frac{1}{R_{i,B}^2}\sum_{j\gamma}\frac{\partial(R_{i\mu,B}R_{i\nu,B})}{\partial r_{j\gamma}}\Delta r_{j\gamma}(k)+R_{i\mu,B}R_{i\nu,B}\sum_{j\gamma}\frac{\partial}{\partial r_{j\gamma}}\left(\frac{1}{R_{i,B}^2}\right)\Delta r_{j\gamma}(k)\right]\right\}\\
&=\frac{1}{2}\sum_{iB}\left\{\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu}+(\alpha'_{\parallel,B}-\alpha'_{\perp,B})\left(\frac{R_{i\mu,B}R_{i\nu,B}}{R_{i,B}^2}-\frac{1}{3}\delta_{\mu\nu}\right)\right]\left(-2\frac{\boldsymbol{R}_{i,B}}{R_{i,B}}\cdot\Delta\vec{r}_i(k)\right)\right\}\\
&\quad+\frac{1}{2}\sum_{iB}\left\{(\alpha_{\parallel,B}-\alpha_{\perp,B})\left[-\frac{2}{R_{i,B}^2}\left(R_{i\nu,B}\Delta r_{i\mu}(k)+R_{i\mu,B}\Delta r_{i\nu}(k)\right)+4R_{i\mu,B}R_{i\nu,B}\frac{\boldsymbol{R}_{i,B}}{R_{i,B}^4}\cdot\Delta\vec{r}_i(k)\right]\right\}\text{(see Eqs. S5-S7)}\\
&=-\sum_{iB}\left\{\frac{\boldsymbol{R}_{i,B}}{R_{i,B}}\cdot\Delta\vec{r}_i(k)\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu}+\left(\alpha'_{\parallel,B}-\alpha'_{\perp,B}\right)\left(\frac{R_{i\mu,B}R_{i\nu,B}}{R_{i,B}^2}-\frac{1}{3}\delta_{\mu\nu}\right)\right]\right\}\\
&\quad-\sum_{iB}\left\{\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\left[\frac{R_{i\nu,B}\Delta r_{i\mu}(k)+R_{i\mu,B}\Delta r_{i\nu}(k)}{R_{i,B}}-2\frac{R_{i\mu,B}R_{i\nu,B}}{R_{i,B}^2}\frac{\boldsymbol{R}_{i,B}}{R_{i,B}}\cdot\Delta\vec{r}_i(k)\right]\right\}\\
&=-\sum_{iB}\left\{\hat{\boldsymbol{R}}_{i,B}\cdot\Delta\vec{r}_i(k)\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu}+\left(\alpha'_{\parallel,B}-\alpha'_{\perp,B}\right)\left(\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}-\frac{1}{3}\delta_{\mu\nu}\right)\right]\right\}\\
&\quad-\sum_{iB}\left\{\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\left[\hat{R}_{i\mu,B}\Delta r_{i\nu}(k)+\hat{R}_{i\nu,B}\Delta r_{i\mu}(k)-2\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}\left(\hat{\boldsymbol{R}}_{i,B}\cdot\Delta\vec{r}_i(k)\right)\right]\right\},
\end{aligned}\tag{S8}$$

where $\hat{\boldsymbol{R}}_{i,B}=\dfrac{\boldsymbol{R}_{i,B}}{R_{i,B}}$ is the equilibrium-configuration bond vector normalized to unity, $\hat{R}_{i\mu,B}$ is the μ (x, y or z) component of the normalized bond vector, and $R_{i,B}$ is the bond length in equilibrium.

For an interlayer shear mode vibrating along the x direction, only the x component of $\Delta\vec{r}_i(k)$ can be non-zero, which yields

$$\begin{aligned}
\Delta\alpha_{\mu\nu} &= -\sum_{iB}\left\{\hat{R}_{ix,B}\Delta r_{ix}\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu}+\left(\alpha'_{\parallel,B}-\alpha'_{\perp,B}\right)\left(\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}-\frac{1}{3}\delta_{\mu\nu}\right)\right]\right\}\\
&\quad-\sum_{iB}\left\{\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\left[\hat{R}_{i\mu,B}\Delta r_{ix}\delta_{\nu x}+\hat{R}_{i\nu,B}\Delta r_{ix}\delta_{\mu x}-2\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}\left(\hat{R}_{ix,B}\Delta r_{ix}\right)\right]\right\}\\
&= -\sum_{iB}\left\{\hat{R}_{ix,B}\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu}+\left(\alpha'_{\parallel,B}-\alpha'_{\perp,B}\right)\left(\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}-\frac{1}{3}\delta_{\mu\nu}\right)\right]\right.\\
&\qquad\left.+\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\left[\hat{R}_{i\mu,B}\delta_{\nu x}+\hat{R}_{i\nu,B}\delta_{\mu x}-2\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}\hat{R}_{ix,B}\right]\right\}\Delta r_{ix}.
\end{aligned}\tag{S9}$$

As discussed in the main text, for an interlayer vibrational mode in 2D materials, each layer vibrates as an almost rigid body and thus it can be simplified as a single object, where the structural details of each layer can be omitted. Subsequently, here i indicates the index of an entire layer instead of any atom within it, and B indicates a bond connecting from layer i to a neighboring layer i' in equilibrium. Recalling in the main text that the change of the polarizability by the shear vibration is $\Delta\alpha = \sum_i \alpha'_{ix}\Delta r_{ix}$, and α'_{ix} and $\Delta\alpha$ are second-rank tensors. Thus we have $\Delta\alpha_{\mu\nu} = \sum_i \alpha'_{ix,\mu\nu}\Delta r_{ix}$. Comparing this equation with the above Eq. S9, we arrive at

$$\begin{aligned}\alpha'_{ix,\mu\nu} = -\sum_B \Bigg\{ \hat{R}_{ix,B}\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu} + \left(\alpha'_{\parallel,B}-\alpha'_{\perp,B}\right)\left(\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}-\frac{1}{3}\delta_{\mu\nu}\right)\right] \\ + \frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\left[\hat{R}_{i\mu,B}\delta_{\nu x}+\hat{R}_{i\nu,B}\delta_{\mu x}-2\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}\hat{R}_{ix,B}\right]\Bigg\}. \end{aligned} \tag{S10}$$

Similarly for an interlayer breathing mode, only the z component of $\Delta\vec{r}_i(k)$ can be non-zero, and thus we obtain

$$\begin{aligned}\Delta\alpha_{\mu\nu} = -\sum_{iB} \Bigg\{ \hat{R}_{iz,B}\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu} + \left(\alpha'_{\parallel,B}-\alpha'_{\perp,B}\right)\left(\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}-\frac{1}{3}\delta_{\mu\nu}\right)\right] \\ + \frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\left[\hat{R}_{i\mu,B}\delta_{\nu z}+\hat{R}_{i\nu,B}\delta_{\mu z}-2\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}\hat{R}_{iz,B}\right]\Bigg\}\Delta r_{iz}. \end{aligned} \tag{S11}$$

Again recalling in the main text that the change of the polarizability by the breathing vibration is $\Delta\alpha_{\mu\nu} = \sum_i \alpha'_{iz,\mu\nu}\Delta r_{iz}$. Comparing this equation with the above Eq. S11, we arrive at

$$\begin{aligned}\alpha'_{iz,\mu\nu} = -\sum_B \Bigg\{ \hat{R}_{iz,B}\left[\frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\delta_{\mu\nu} + \left(\alpha'_{\parallel,B}-\alpha'_{\perp,B}\right)\left(\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}-\frac{1}{3}\delta_{\mu\nu}\right)\right] \\ + \frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\left[\hat{R}_{i\mu,B}\delta_{\nu z}+\hat{R}_{i\nu,B}\delta_{\mu z}-2\hat{R}_{i\mu,B}\hat{R}_{i\nu,B}\hat{R}_{iz,B}\right]\Bigg\}. \end{aligned} \tag{S12}$$

Eq. S10 and Eq. S12 suggest that α'_{ix} or α'_{iz}, the derivative of the system's polarizability with respect to the layer i's displacement along the x or z direction, can be determined by the interlayer bond (length and direction), and bond polarizabilities.

According to Eq. S1, for the commonly used parallel polarization set-up in the backscattering geometry $z(xx)\bar{z}$, only the xx components of the tensors need to be considered (i.e., $\mu = \nu = x$). Consequently, we have

$$\begin{aligned}\alpha'_{ix,xx} &= -\sum_B \Bigg\{ \frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\hat{R}_{ix,B} + (\alpha'_{\parallel,B}-\alpha'_{\perp,B})\hat{R}^3_{ix,B} - \frac{\alpha'_{\parallel,B}-\alpha'_{\perp,B}}{3}\hat{R}_{ix,B} \\ &\qquad +2\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\hat{R}_{ix,B} - 2\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\hat{R}^3_{ix,B}\Bigg\} \\ &= -\sum_B \Bigg\{ \frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3} + (\alpha'_{\parallel,B}-\alpha'_{\perp,B})\hat{R}^2_{ix,B} - \frac{\alpha'_{\parallel,B}-\alpha'_{\perp,B}}{3} + 2\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}} - 2\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\hat{R}^2_{ix,B}\Bigg\}\hat{R}_{ix,B} \\ &= \sum_B C_{i,B}\hat{R}_{ix,B}, \end{aligned} \tag{S13}$$

and

$$\begin{aligned}
\alpha'_{iz,xx} &= -\sum_B \left\{ \frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3}\hat{R}_{iz,B} + (\alpha'_{\parallel,B}-\alpha'_{\perp,B})\hat{R}^2_{ix,B}\hat{R}_{iz,B} - \frac{\alpha'_{\parallel,B}-\alpha'_{\perp,B}}{3}\hat{R}_{iz,B} - 2\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\hat{R}^2_{ix,B}\hat{R}_{iz,B} \right\} \\
&= -\sum_B \left\{ \frac{\alpha'_{\parallel,B}+2\alpha'_{\perp,B}}{3} + (\alpha'_{\parallel,B}-\alpha'_{\perp,B})\hat{R}^2_{ix,B} - \frac{\alpha'_{\parallel,B}-\alpha'_{\perp,B}}{3} - 2\frac{\alpha_{\parallel,B}-\alpha_{\perp,B}}{R_{i,B}}\hat{R}^2_{ix,B} \right\} \hat{R}_{iz,B} \\
&= \sum_B C^*_{i,B}\hat{R}_{iz,B},
\end{aligned} \tag{S14}$$

where the coefficients $C_{i,B}$ and $C^*_{i,B}$ are related to the properties of the interlayer bond B connecting from layer i to a neighboring layer i', such as the interlayer bond length and its x component, and the interlayer bond polarizabilities and their radial derivatives.

It follows that the change of the polarizability is

$$\Delta\alpha_{xx} = \sum_i \alpha'_{ix,xx}\Delta r_{ix} \tag{S15}$$

by the shear vibrations and

$$\Delta\alpha_{xx} = \sum_i \alpha'_{iz,xx}\Delta r_{iz} \tag{S16}$$

by the breathing vibrations.

S2 The interlayer bonds and polarizability derivatives of each layer

As shown in Figure S2, except that layer 1 and layer N have only one interlayer bond, other interior layers i have two interlayer bonds: one with the layer above $i-1$ and the other one with the layer below $i+1$. For an interior layer i, the x components of these two normalized bond vectors assume a relation $\hat{R}_{ix,i-1} = \hat{R}_{ix,i+1}$ in (a) AB or AA$'$ stacking, while $\hat{R}_{ix,i-1} = -\hat{R}_{ix,i+1}$ in (b) ABC stacking. In addition, regardless of the stacking type, for layer i and its neighboring layer j ($i-1$ or $i+1$), there is a general relation $\hat{R}_{ix,j} = -\hat{R}_{jx,i}$.

For AB or AA$'$ stacking in Figure S2a, taking 6L as an example, $\hat{R}_{1x,2} = \sin\theta, \hat{R}_{2x,1} = \hat{R}_{2x,3} = -\sin\theta, \hat{R}_{3x,2} = \hat{R}_{3x,4} = \sin\theta, \hat{R}_{4x,3} = \hat{R}_{4x,5} = -\sin\theta, \hat{R}_{5x,4} = \hat{R}_{5x,6} = \sin\theta, \hat{R}_{6x,5} = -\sin\theta$. Thus the polarizability derivative with respect to the layer i's displacement along the x direction is $\alpha'_{1x,xx} = C\hat{R}_{1x,2} = C\sin\theta = \beta, \alpha'_{2x,xx} = C(\hat{R}_{2x,1}+\hat{R}_{2x,3}) = -2C\sin\theta = -2\beta, \alpha'_{3x,xx} = C(\hat{R}_{3x,2}+\hat{R}_{3x,4}) = 2C\sin\theta = 2\beta, \alpha'_{4x,xx} = -2\beta, \alpha'_{5x,xx} = 2\beta, \alpha'_{6x,xx} = C\hat{R}_{6x,5} = -C\sin\theta = -\beta$. Note that $C = C(\mathrm{AB})$ or $C = C(\mathrm{AA'})$, the coefficient related to the interlayer bond polarizability and its derivatives in AB or AA$'$ stacking, respectively. Taking 7L as an example, the interlayer bond vectors are not changed for layer 1 to layer 5, but layer 6 has $\hat{R}_{6x,5} = \hat{R}_{6x,7} = -\sin\theta$, while layer 7 has $\hat{R}_{7x,6} = \sin\theta$. Thus we have $\alpha'_{1x,xx} = \beta, \alpha'_{2x,xx} = -2\beta, \alpha'_{3x,xx} = 2\beta, \alpha'_{4x,xx} = -2\beta, \alpha'_{5x,xx} = 2\beta, \alpha'_{6x,xx} = C(\hat{R}_{6x,5}+\hat{R}_{6x,7}) = -2C\sin\theta = -2\beta, \alpha'_{7x,xx} = C\hat{R}_{7x,6} = C\sin\theta = \beta$. In general, for AB or AA$'$ stacking, due to $\hat{R}_{ix,i-1} = \hat{R}_{ix,i+1}$, $\alpha'_{1x,xx} = \beta$, $\alpha'_{Nx,xx} = \beta$ for odd N or $\alpha'_{Nx,xx} = -\beta$ for even N, and $\alpha'_{2x,xx} = -2\beta, \alpha'_{3x,xx} = 2\beta, \alpha'_{4x,xx} = -2\beta, \alpha'_{5x,xx} = 2\beta, \ldots$, where there is a repeated pattern of $-2\beta, 2\beta$ for the interior layers. Here $\beta = \beta_1$ for AA$'$ stacking, while $\beta = \beta_2$ for AB stacking.

For ABC stacking in Figure S2b, due to $\hat{R}_{ix,i-1} = -\hat{R}_{ix,i+1}$, for an interior layer i, $\alpha'_{ix,xx} = C(\hat{R}_{ix,i-1} + \hat{R}_{ix,i+1}) = 0$, while for layer 1 and layer N, $\alpha'_{1x,xx} = C\hat{R}_{1x,2} = C\sin\theta = \beta$ and $\alpha'_{Nx,xx} = C\hat{R}_{Nx,N-1} = -C\sin\theta = -\beta$. Here $\beta = \beta_2$ for ABC stacking.

For AA$'$B$'$B stacking in Figure S2c, the periodicity corresponds to every four layers, and AA$'$ and AB stackings (red and blue colors) alternate. The x components of normalized interlayer bond vectors are $\hat{R}_{1x,2} =$

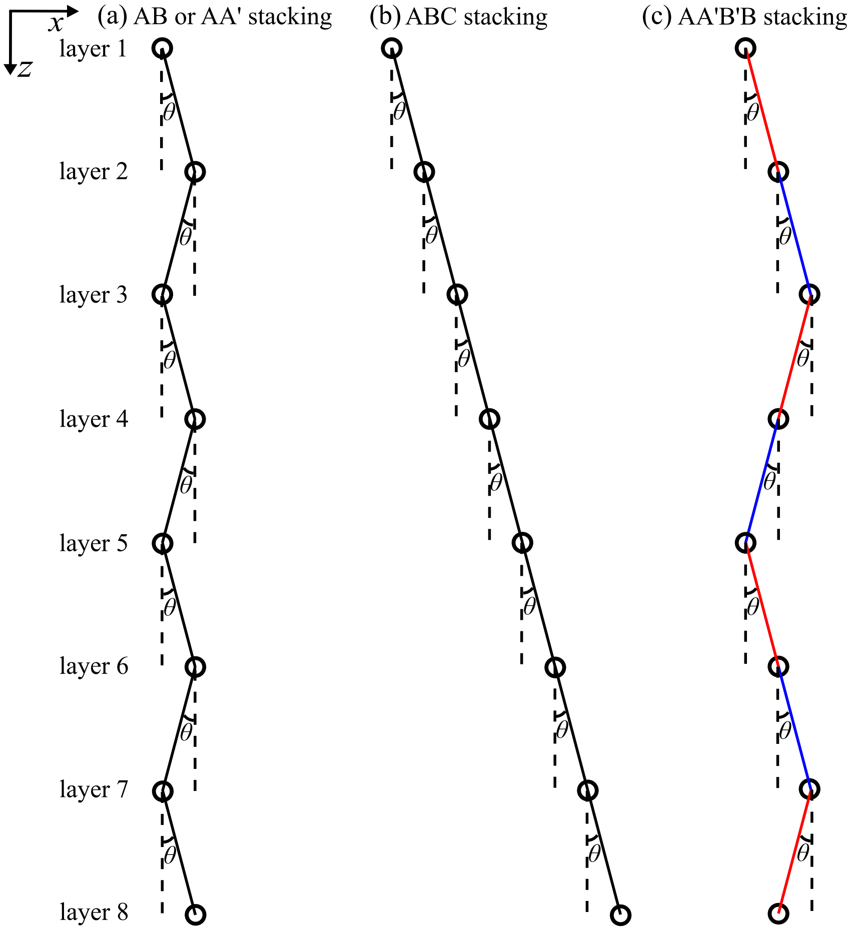

Figure S2 Schematic of the interlayer bonds in N-layer for (a) AB or AA$'$ stacking, (b) ABC stacking, and (c) AA$'$B$'$B stacking types. Each layer is simplified as a single object for interlayer vibrations. AB or AA$'$ stacking repeats every two layer, ABC stacking repeats every three layers, and AA$'$B$'$B stacking repeats every four layers. In (c) AA$'$B$'$B stacking, the interlayer bonds corresponding to AA$'$ and AB stackings are differentiated by red and blue colors, respectively, indicating that AA$'$ and AB stackings alternate.

$\sin\theta, \hat{R}_{2x,1} = -\hat{R}_{2x,3} = -\sin\theta, \hat{R}_{3x,2} = \hat{R}_{3x,4} = -\sin\theta, \hat{R}_{4x,3} = -\hat{R}_{4x,5} = \sin\theta, \hat{R}_{5x,4} = \hat{R}_{5x,6} = \sin\theta, \hat{R}_{6x,5} = -\hat{R}_{6x,7} = -\sin\theta, \hat{R}_{7x,6} = \hat{R}_{7x,8} = -\sin\theta, \hat{R}_{8x,7} = -\hat{R}_{8x,9} = \sin\theta, \ldots$, where an interior layer i has the same interlayer bond vectors to layer $i+4$. Thus

$$\begin{aligned}
\alpha'_{1x,xx} &= C_{1,2}\hat{R}_{1x,2} = C(\mathrm{AA}')\sin\theta = \beta_1, \\
\alpha'_{2x,xx} &= C_{2,1}\hat{R}_{2x,1} + C_{2,3}\hat{R}_{2x,3} = -C(\mathrm{AA}')\sin\theta + C(\mathrm{AB})\sin\theta = -\beta_1 + \beta_2, \\
\alpha'_{3x,xx} &= C_{3,2}\hat{R}_{3x,2} + C_{3,4}\hat{R}_{3x,4} = -C(\mathrm{AB})\sin\theta - C(\mathrm{AA}')\sin\theta = -\beta_2 - \beta_1, \\
\alpha'_{4x,xx} &= C_{4,3}\hat{R}_{4x,3} + C_{4,5}\hat{R}_{4x,5} = C(\mathrm{AA}')\sin\theta - C(\mathrm{AB})\sin\theta = \beta_1 - \beta_2, \\
\alpha'_{5x,xx} &= C_{5,4}\hat{R}_{5x,4} + C_{5,6}\hat{R}_{5x,6} = C(\mathrm{AB})\sin\theta + C(\mathrm{AA}')\sin\theta = \beta_2 + \beta_1, \\
\alpha'_{6x,xx} &= C_{6,5}\hat{R}_{6x,5} + C_{6,7}\hat{R}_{6x,7} = -C(\mathrm{AA}')\sin\theta + C(\mathrm{AB})\sin\theta = -\beta_1 + \beta_2, \\
\alpha'_{7x,xx} &= C_{7,6}\hat{R}_{7x,6} + C_{7,8}\hat{R}_{7x,8} = -C(\mathrm{AB})\sin\theta - C(\mathrm{AA}')\sin\theta = -\beta_2 - \beta_1, \\
\alpha'_{8x,xx} &= C_{8,7}\hat{R}_{8x,7} + C_{8,9}\hat{R}_{8x,9} = C(\mathrm{AA}')\sin\theta - C(\mathrm{AB})\sin\theta = \beta_1 - \beta_2, \\
&\vdots \\
\alpha'_{Nx,xx} &= \beta_1(\text{if } N = 4m) \text{ or } \beta_2(\text{if } N = 4m+1) \text{ or } -\beta_1(\text{if } N = 4m+2) \text{ or } -\beta_2(\text{if } N = 4m+3),
\end{aligned} \tag{S17}$$

where m is an integer, $C(\mathrm{AA}')\sin\theta = \beta_1$, and $C(\mathrm{AB})\sin\theta = \beta_2$. Note that for an interior layer i, $\alpha'_{ix,xx} = -\alpha'_{(i+2)x,xx}$, and thus $\alpha'_{ix,xx} = \alpha'_{(i+4)x,xx}$.

Turing to the z direction (Figure S2), for an interior layer i, the z components of the two normalized interlayer bond vectors always assume a relation $\hat{R}_{iz,i-1} = -\hat{R}_{iz,i+1}$ regardless of the in-plane stacking details. In addition, regardless of the stacking type, for layer i and its neighboring layer j ($i-1$ or $i+1$), there is a general relation $\hat{R}_{iz,j} = -\hat{R}_{jz,i}$. In AB or AA′ or ABC stacking, for an interior layer i, the polarizability derivative with respect to its displacement along the z direction is $\alpha'_{iz,xx} = C^*(\hat{R}_{iz,i-1} + \hat{R}_{iz,i+1}) = 0$, while for layer 1 and layer N, $\alpha'_{1z,xx} = C^*\hat{R}_{1z,2} = C^*\cos\theta = \gamma$ and $\alpha'_{Nz,xx} = C^*\hat{R}_{Nz,N-1} = -C^*\cos\theta = -\gamma$. Here $\gamma = \gamma_1$ for AA′ stacking, while $\gamma = \gamma_2$ for AB or ABC stacking.

However, again for AA′B′B stacking, the situation is more complicated due to the mixture of AA′ and AB stackings. In specific,

$$\begin{aligned}
\alpha'_{1z,xx} &= C^*_{1,2}\hat{R}_{1z,2} = C(\mathrm{AA}')^*\cos\theta = \gamma_1,\\
\alpha'_{2z,xx} &= C^*_{2,1}\hat{R}_{2z,1} + C^*_{2,3}\hat{R}_{2z,3} = -C(\mathrm{AA}')^*\cos\theta + C(\mathrm{AB})^*\cos\theta = -\gamma_1 + \gamma_2,\\
\alpha'_{3z,xx} &= C^*_{3,2}\hat{R}_{3z,2} + C^*_{3,4}\hat{R}_{3z,4} = -C(\mathrm{AB})^*\cos\theta + C(\mathrm{AA}')^*\cos\theta = -\gamma_2 + \gamma_1,\\
\alpha'_{4z,xx} &= C^*_{4,3}\hat{R}_{4z,3} + C^*_{4,5}\hat{R}_{4z,5} = -C(\mathrm{AA}')^*\cos\theta + C(\mathrm{AB})^*\cos\theta = -\gamma_1 + \gamma_2,\\
\alpha'_{5z,xx} &= C^*_{5,4}\hat{R}_{5z,4} + C^*_{5,6}\hat{R}_{5z,6} = -C(\mathrm{AB})^*\cos\theta + C(\mathrm{AA}')^*\cos\theta = -\gamma_2 + \gamma_1,\\
&\ \vdots\\
\alpha'_{Nz,xx} &= -\gamma_1(\text{if } N = 2m) \text{ or } -\gamma_2(\text{if } N = 2m+1),
\end{aligned} \tag{S18}$$

where m is an integer, $C(\mathrm{AA}')^*\cos\theta = \gamma_1$, and $C(\mathrm{AB})^*\cos\theta = \gamma_2$. Note that for an interior layer i, $\alpha'_{iz,xx} = -\alpha'_{(i+1)z,xx}$ and thus $\alpha'_{iz,xx} = \alpha'_{(i+2)z,xx}$.

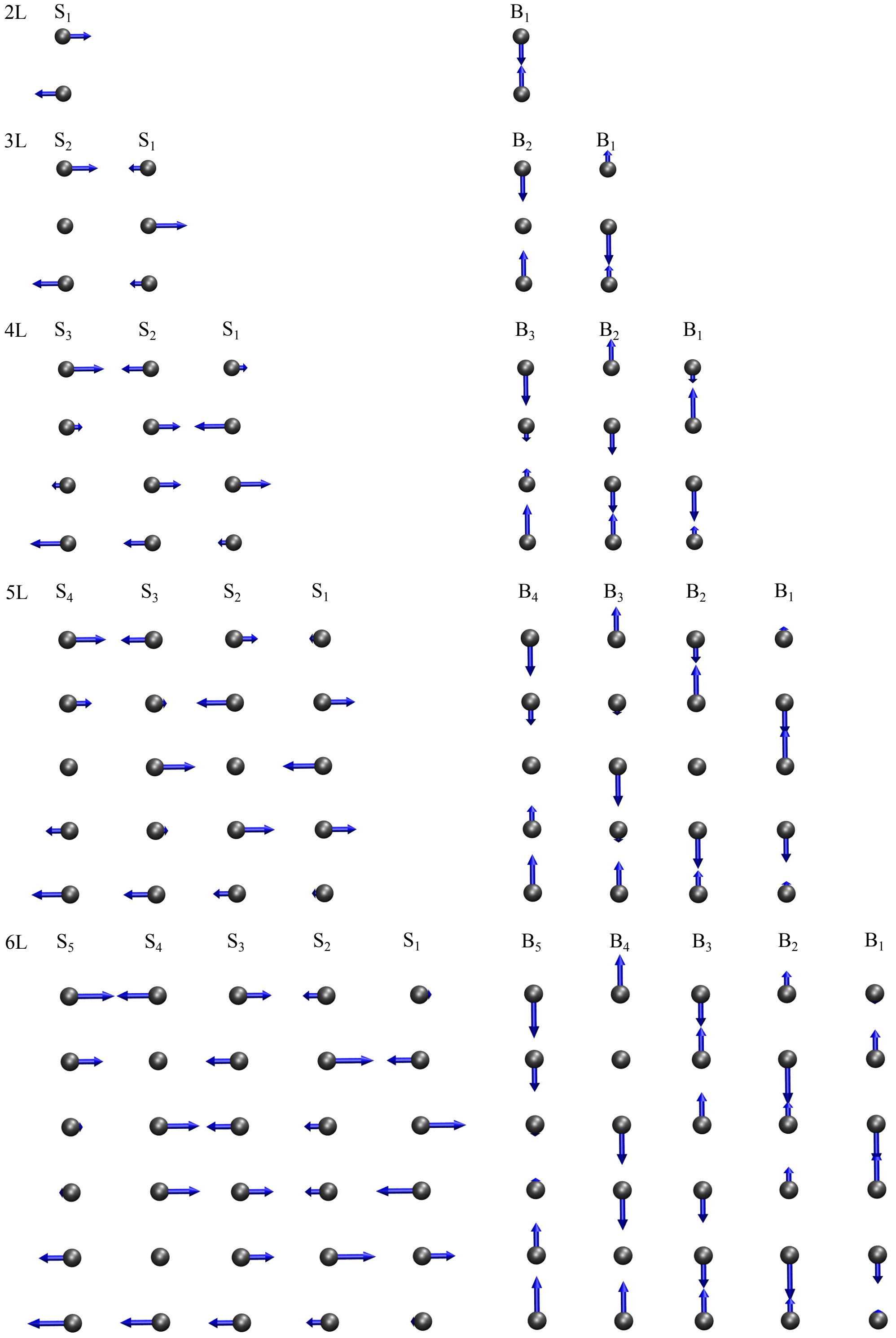

Figure S3 Schematic of the vibrations of the interlayer shear (S) and breathing (B) modes from 2L to 6L, derived from the linear chain model. For these interlayer vibrations, each layer is treated as a single object (the gray sphere), and the blue arrows indicate both the direction and magnitude of the vibrations of each layer. For NL, there are $N-1$ S and B modes, where S_1 (B_1) is the highest-frequency S (B) mode, while S_{N-1} (B_{N-1}) is the lowest-frequency S (B) mode.

S3 Parameter fitting for trilayer $MoSe_2$

As discussed in the main text, for trilayer $MoSe_2$ in $AA'B'$ stacking, the polarizability change by the shear vibrations can be derived as follows:

$$\Delta\alpha_{xx}(AA'B', S_2) = \sqrt{0.5}(\beta_1 + \beta_2); \qquad \Delta\alpha_{xx}(AA'B', S_1) = \sqrt{1.5}(\beta_1 - \beta_2).$$

For 3L $MoSe_2$, the frequencies of the S_2 and S_1 modes are $\omega_2 \approx 14$ cm^{-1} and $\omega_1 \approx 23$ cm^{-1}, respectively. In $AA'B'$ stacking, we have $I(S_2) = \frac{n_2+1}{\omega_2}|\Delta\alpha_{xx}(AA'B', S_2)|^2 = 0.5\frac{n_2+1}{\omega_2}|\beta_1+\beta_2|^2 \approx 0.59|\beta_1+\beta_2|^2$ and $I(S_1) = \frac{n_1+1}{\omega_1}|\Delta\alpha_{xx}(AA'B', S_1)|^2 = 1.5\frac{n_1+1}{\omega_1}|\beta_1-\beta_2|^2 \approx 0.61|\beta_1-\beta_2|^2$. Here $n_i = (e^{\hbar\omega_i/k_BT} - 1)^{-1}$ is the Bose-Einstein distribution of phonon occupation at room temperature $T = 300$K. Based on the experimental Raman data of bilayer $MoSe_2$,[6] we know $|\beta_1|/|\beta_2| = 2.32$ as mentioned in the main text.

If β_1 and β_2 are assumed to be real variables as in the common non-resonant Raman modeling, then $\beta_1 = 2.32\beta_2$ or $\beta_1 = -2.32\beta_2$. For the former case, we have $I(S_2) = 0.59|\beta_1+\beta_2|^2 = 0.59|2.32\beta_2+\beta_2|^2 = 6.50|\beta_2|^2$, and $I(S_1) = 0.61|\beta_1-\beta_2|^2 = 0.61|2.32\beta_2-\beta_2|^2 = 1.06|\beta_2|^2$, thereby giving $I(S_2)/I(S_1) = 6.13$; for the latter case, we have $I(S_2) = 0.59|\beta_1+\beta_2|^2 = 0.59|-2.32\beta_2+\beta_2|^2 = 1.03|\beta_2|^2$, and $I(S_1) = 0.61|\beta_1-\beta_2|^2 = 0.61|-2.32\beta_2-\beta_2|^2 = 6.72|\beta_2|^2$, thereby giving $I(S_2)/I(S_1) = 0.15$. Both cases yield very unequal intensities of the S_2 and S_1 modes, which are consistent with first-principles non-resonant Raman calculations in Ref. 6.

However, the S_2 and S_1 modes exhibited nearly equal intensities in the experimental resonant Raman scattering.[6] In reality, the polarizability (or dielectric function) has both real and imaginary parts due to the light absorption in experimental resonant Raman scattering.[7,8] Thus β_1 and β_2 are complex variables: $\beta_1 = |\beta_1|e^{i\phi_1}; \beta_2 = |\beta_2|e^{i\phi_2}$, where ϕ_1 and ϕ_2 are their phase angles, respectively. To have $I(S_2) = I(S_1)$, we need $0.59|\beta_1+\beta_2|^2 = 0.61|\beta_1-\beta_2|^2$, which is

$$\begin{aligned}
&|\beta_1+\beta_2|^2 = 1.034|\beta_1-\beta_2|^2 \longrightarrow \\
&|\beta_1|^2 + |\beta_2|^2 + \beta_1\beta_2^* + \beta_1^*\beta_2 = 1.034\left(|\beta_1|^2 + |\beta_2|^2 - \beta_1\beta_2^* - \beta_1^*\beta_2\right) \longrightarrow \\
&2.034(\beta_1\beta_2^* + \beta_1^*\beta_2) = 0.034\left(|\beta_1|^2 + |\beta_2|^2\right) \longrightarrow \\
&2.034|\beta_1||\beta_2|\left(e^{i(\phi_1-\phi_2)} + e^{-i(\phi_1-\phi_2)}\right) = 0.034\left(|\beta_1|^2 + |\beta_2|^2\right) \longrightarrow \\
&4.068|\beta_1||\beta_2|\cos(\phi_1-\phi_2) = 0.034\left(|\beta_1|^2 + |\beta_2|^2\right) \longrightarrow \\
&\cos(\phi_1-\phi_2) = 0.008\frac{|\beta_1|^2 + |\beta_2|^2}{|\beta_1||\beta_2|}
\end{aligned} \tag{S19}$$

With $|\beta_1| = 2.32|\beta_2|$, we arrive at $\cos(\phi_1-\phi_2) = 0.022$, which yields $|\phi_1-\phi_2| \approx 88.74°$. This suggests that for AA' and AB stackings, their complex interlayer bond polarizabilities and derivatives not only have different magnitudes, but also have different phase angles in the resonant Raman scattering. Here we assume $|\beta_1| = 2.32$ and $\phi_1 = 118.74°$, while $|\beta_2| = 1.00$ and $\phi_2 = 30.00°$ without loss of generality. These parameters give rise to nearly equal intensities between the S_2 and S_1 modes for trilayer $MoSe_2$ in $AA'B'$ stacking.

References

[1] X. Luo, X. Lu, C. Cong, T. Yu, Q. Xiong and S. Y. Quek, *Sci. Rep.*, 2015, **5**, 14565.

[2] D. Snoke and M. Cardona, *Solid State Comm.*, 1993, **87**, 121–126.

[3] S. Guha, J. Menendez, J. Page and G. Adams, *Phys. Rev. B*, 1996, **53**, 13106.

[4] R. Saito, M. Furukawa, G. Dresselhaus and M. Dresselhaus, *J. Phys.Condens. Matter*, 2010, **22**, 334203.

[5] L. Liang and V. Meunier, *Nanoscale*, 2014, **6**, 5394.

[6] A. A. Puretzky, L. Liang, X. Li, K. Xiao, K. Wang, M. Mahjouri-Samani, L. Basile, J. C. Idrobo, B. G. Sumpter, V. Meunier and D. B. Geohegan, *ACS Nano*, 2015, **9**, 6333–6342.

[7] H. B. Ribeiro, M. A. Pimenta, C. J. De Matos, R. L. Moreira, A. S. Rodin, J. D. Zapata, E. A. De Souza and A. H. Castro Neto, *ACS Nano*, 2015, **9**, 4270–4276.

[8] X. Ling, S. Huang, E. H. Hasdeo, L. Liang, W. M. Parkin, Y. Tatsumi, A. R. Nugraha, A. A. Puretzky, P. M. Das, B. G. Sumpter *et al.*, *Nano Lett.*, 2016, **16**, 2260–2267.