Electronic Supplementary Information
Intrinsic effect of interfacial coupling on the high-frequency intralayer modes in twisted multilayer $MoTe_2$

Yu-Chen Leng,[ab] Miao-Ling Lin,[a] Yu Zhou,[c] Jiang-Bin Wu,[a] Da Meng,[a] Xin Cong,[ab] Hai Li,[c] and Ping-Heng Tan*[abd]

Diatomic Chain Model for out-of-plane intralayer mode in N-layer $MoTe_2$

The schematic diagrams of atomic displacements of the out-of-plane intralayer modes in N-layer $MoTe_2$ (NLM) are obtained based on the diatomic linear chain model (DCM)[1]. Two force constants, α_{mt} and α_{tt} ,are needed to describe the vibration modes in NLM. α_{mt} represents the force constant per unit area between the nearest Mo and Te planes within a $MoTe_2$ layer and α_{tt} represents the force constant per unit area between two nearest Te planes in two adjacent layers. Since the Mo and Te atoms in the A_1'-like and A_2''-like modes vibrate perpendicular to the basal plane, only the perpendicular components of the force constants need to be considered. For simplicity, all the following α_{mt} and α_{tt} represent the perpendicular components of the corresponding force constants. $\alpha_{tt} = 7.91 \times 10^{19} N/m^3$ can be obtained by the frequency of the layer-breathing (LB) modes, as described in main text. α_{mt} can be calculated from the frequency of the A_1' mode in 1LM:
The vibrations of the three atoms in 1LM can be described by the following equations,

$$\begin{cases} m_{\mathrm{Te}}\ddot{U}_{\mathrm{Te1}} = -\alpha_{mt}\ddot{U}_{\mathrm{Te1}} + \alpha_{mt}\ddot{U}_{\mathrm{Mo}} \\ m_{\mathrm{Mo}}\ddot{U}_{\mathrm{Mo}} = \alpha_{mt}\ddot{U}_{\mathrm{Te1}} - 2\alpha_{mt}\ddot{U}_{\mathrm{Mo}} + \alpha_{mt}\ddot{U}_{\mathrm{Te2}} \\ m_{\mathrm{Te}}\ddot{U}_{\mathrm{Te2}} = \alpha_{mt}\ddot{U}_{\mathrm{Mo}} - \alpha_{mt}\ddot{U}_{\mathrm{Te2}} \end{cases} \tag{S1}$$

where m_{Te} and m_{Mo} represent the mass of Te and Mo atoms, respectively. U_{Te} and U_{Mo} represent the displacements of the Te and Mo atoms relative to their corresponding equilibrium positions, respectively, subscripts 1 and 2 are used to distinguish the two different Te atoms in one layer. The solution of the above equations of motion is obtained by using the following substitution,

$$U = u \times e^{-i\omega t} \tag{S2}$$

where u is the amplitude of displacement of one atom, and ω represent its vibration frequency. After the substitution, the following equations are obtained,

$$\begin{cases} \omega^2 u_{\mathrm{Te1}} = -\dfrac{\alpha_{mt}}{m_{\mathrm{Te}}}u_{\mathrm{Te1}} + \dfrac{\alpha_{mt}}{m_{\mathrm{Te}}}u_{\mathrm{Mo}} \\ \omega^2 u_{\mathrm{Mo}} = \dfrac{\alpha_{mt}}{m_{\mathrm{Mo}}}u_{\mathrm{Te1}} - 2\dfrac{\alpha_{mt}}{m_{\mathrm{Mo}}}u_{\mathrm{Mo}} + \dfrac{\alpha_{mt}}{m_{\mathrm{Mo}}}u_{\mathrm{Te2}} \\ \omega^2 u_{\mathrm{Te2}} = \dfrac{\alpha_{mt}}{m_{\mathrm{Te}}}u_{\mathrm{Mo}} - \dfrac{\alpha_{mt}}{m_{\mathrm{Te}}}u_{\mathrm{Te2}} \end{cases} \tag{S3}$$

which can be expressed in a matrix form as,

$$\omega^2 \mathbf{u} = \mathbf{D}\mathbf{u}. \tag{S4}$$

[a] *State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China. E-mail: phtan@semi.ac.cn*
[b] *Center of Materials Science and Optoelectronics Engineering & CAS Center of Excellence in Topological Quantum Computation, University of Chinese Academy of Sciences, Beijing 100049, China.*
[c] *Key Laboratory of Flexible Electronics and Institute of Advanced Materials, Jiangsu National Synergetic Innovation Center for Advanced Materials, Nanjing Tech University, 30 South Puzhu Road, 211816 Nanjing, China.*
[d] *Beijing Academy of Quantum Information Science, Beijing 100193, China.*

This equation is equivalent to

$$2\pi^2 c^2 \omega^2 \mathbf{u}' = \mathbf{D}\mathbf{u}' \tag{S5}$$

where c = 3×10^8 m/s is the speed of light, $\mathbf{u}'$ is the column vector of displacement and $\mathbf{D}$ is a force constant matrix:

$$\mathbf{D} = \begin{pmatrix} -\frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} & \frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} & 0 \\ \frac{\alpha_{mt}}{\mu_{\mathrm{Mo}}} & -2\frac{\alpha_{mt}}{\mu_{\mathrm{Mo}}} & \frac{\alpha_{mt}}{\mu_{\mathrm{Mo}}} \\ 0 & \frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} & -\frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} \end{pmatrix}. \tag{S6}$$

Here, $\mu_{\mathrm{Te}} = 2.03 \times 10^{-6}$ kg/m^2 and $\mu_{\mathrm{Mo}} = 1.53 \times 10^{-6}$ kg/m^2 are the mass of Te and Mo atoms per unit area, respectively. For the A_1' mode of 1LM, the Mo atom do not vibrate, giving $\omega(A_1') = \frac{1}{\sqrt{2}\pi c}\sqrt{\frac{\alpha_{mt}}{\mu_{\mathrm{Te}}}}$.[1] We measure $\omega(A_1') \sim 172.8$ cm^{-1} in 1LM. This gives $\alpha_{mt} = 2.15 \times 10^{21}$ N/m^3.

For *N*LM, the corresponding force constant matrix can be given by a similar method. For example, the force constant matrix D for 2LM can be written as:

$$\mathbf{D} = \begin{pmatrix} -\frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} & \frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} & 0 & 0 & 0 & 0 \\ \frac{\alpha_{mt}}{\mu_{\mathrm{Mo}}} & -2\frac{\alpha_{mt}}{\mu_{\mathrm{Mo}}} & \frac{\alpha_{mt}}{\mu_{\mathrm{Mo}}} & 0 & 0 & 0 \\ 0 & \frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} & -\frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} - \frac{\alpha_{tt}}{\mu_{\mathrm{Te}}} & \frac{\alpha_{tt}}{\mu_{\mathrm{Te}}} & 0 & 0 \\ 0 & 0 & \frac{\alpha_{tt}}{\mu_{\mathrm{Te}}} & -\frac{\alpha_{tt}}{\mu_{\mathrm{Te}}} - \frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} & \frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} & 0 \\ 0 & 0 & 0 & \frac{\alpha_{mt}}{\mu_{\mathrm{Mo}}} & -2\frac{\alpha_{mt}}{\mu_{\mathrm{Mo}}} & \frac{\alpha_{mt}}{\mu_{\mathrm{Mo}}} \\ 0 & 0 & 0 & 0 & \frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} & -\frac{\alpha_{mt}}{\mu_{\mathrm{Te}}} \end{pmatrix}, \tag{S7}$$

Then the displacement of each atom in A_1'-like and A_2''-like modes can be obtained by solving $\mathbf{u}$ in Eq. (S5). The results are summarized in Fig.S1 and S2.

Notes and references

1 X. Zhang, W.-P. Han, J.-B. Wu, S. Milana, Y. Lu, Q.-Q. Li, A. C. Ferrari and P.-H. Tan, *Phys. Rev. B*, 2013, **87**, 115413.

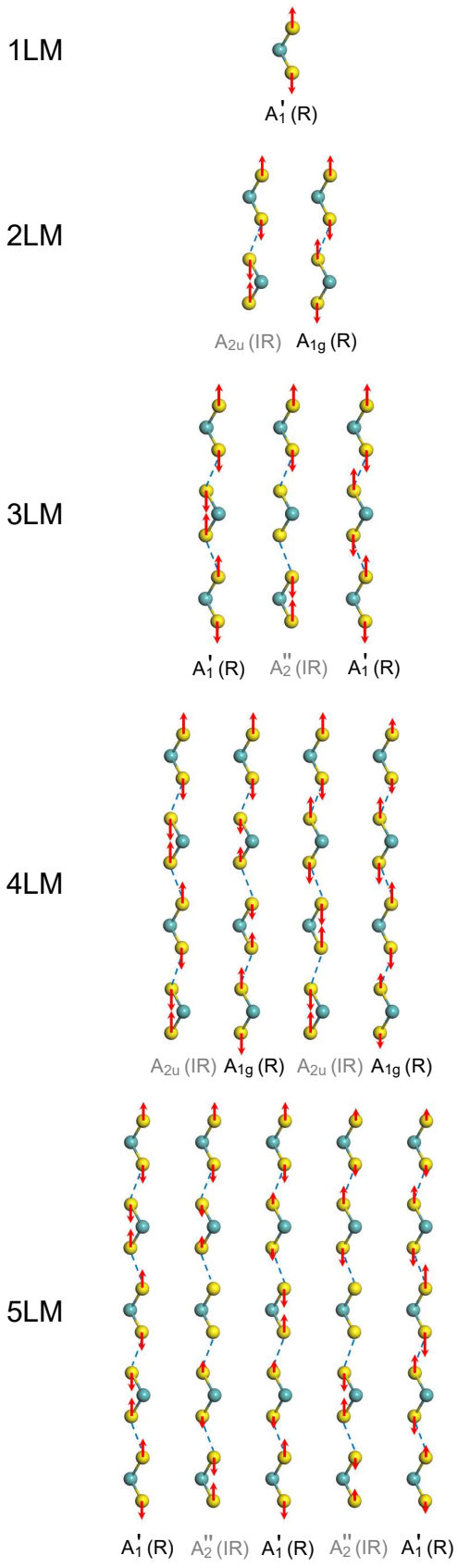

Fig. S1 Calculated atom displacements for the A_1'-like modes($\sim$172 cm^{-1}) in 1LM-5LM by DCM. The frequencies of the modes in each layer increase from left to right. The irreducible representation of each modes with Raman activity(black) and infrared activity(gray) are indicated. The directions and lengths of the arrows represent the directions and amplitudes of the atomic vibration, respectively.

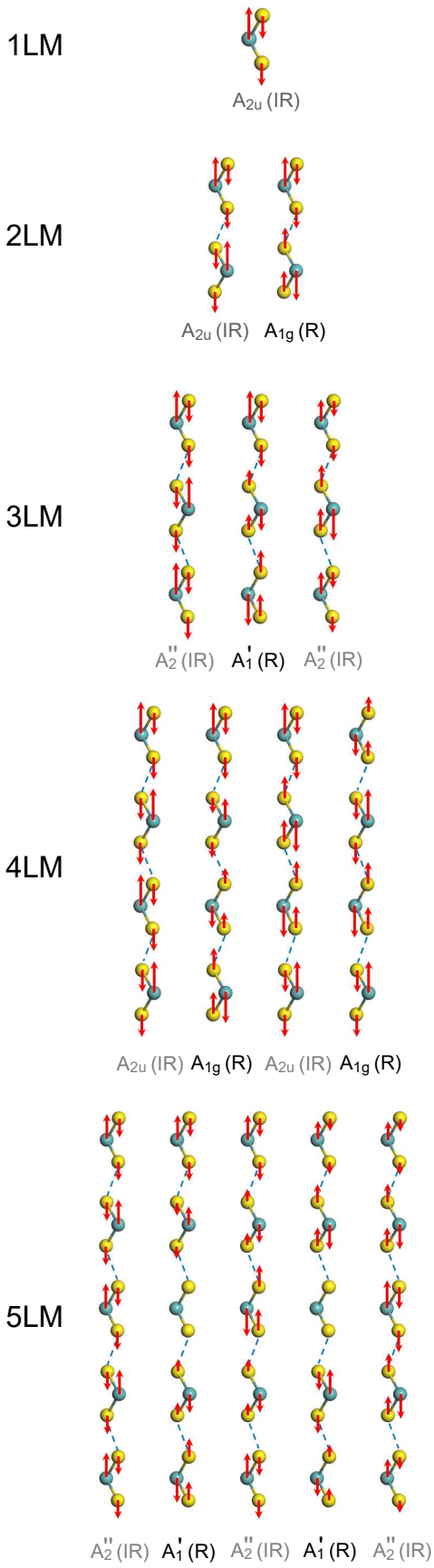

Fig. S2 Calculated atom displacements for the A_2''-like modes($\sim$290 cm^{-1}) in 1LM-5LM by DCM. The frequencies of the modes in each layer increase from left to right. The irreducible representation of each modes with Raman activity(black) and infrared activity(gray) are indicated. The directions and lengths of the arrows represent the directions and amplitudes of the atomic vibration, respectively.

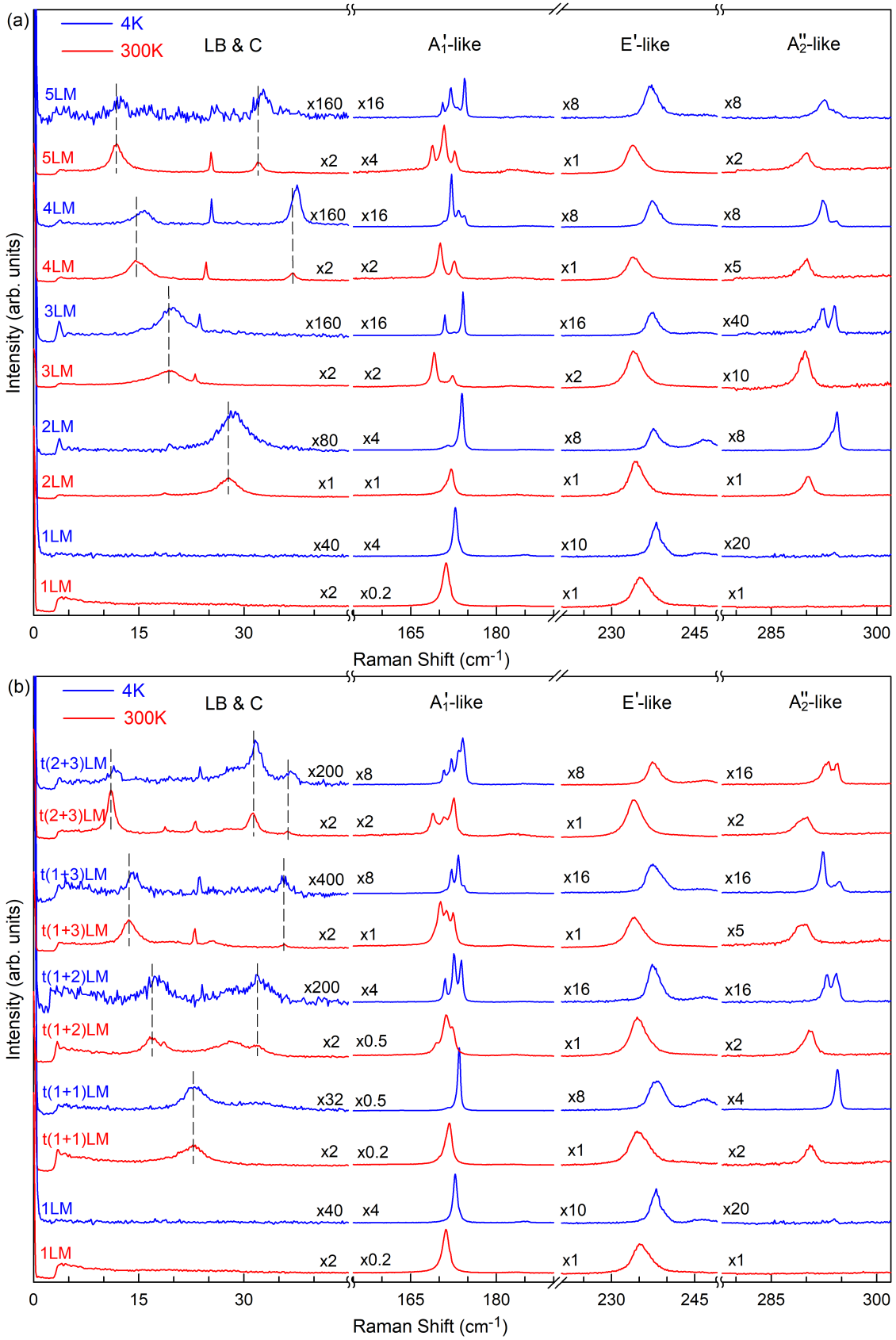

Fig. S3 The Raman spectra of interlayer and intralayer modes in (a) NLM and (b) $t(m+n)$LM at 4K (blue) and 300K (red). Raman spectra are offset for clarity.

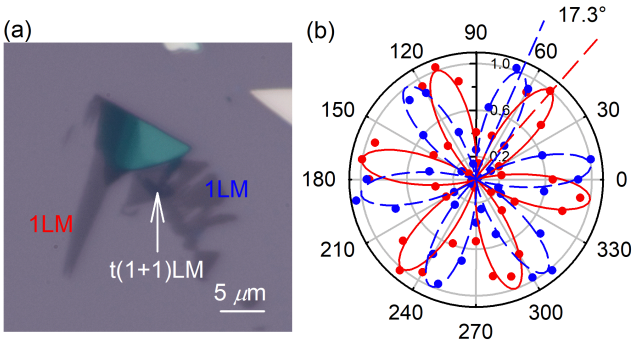

Fig. S4 (a) Optical image of the t(1+1)LM. (b) The crystallographic-orientation dependent SHG intensity of two 1LM constituents (blue circle and red circle).

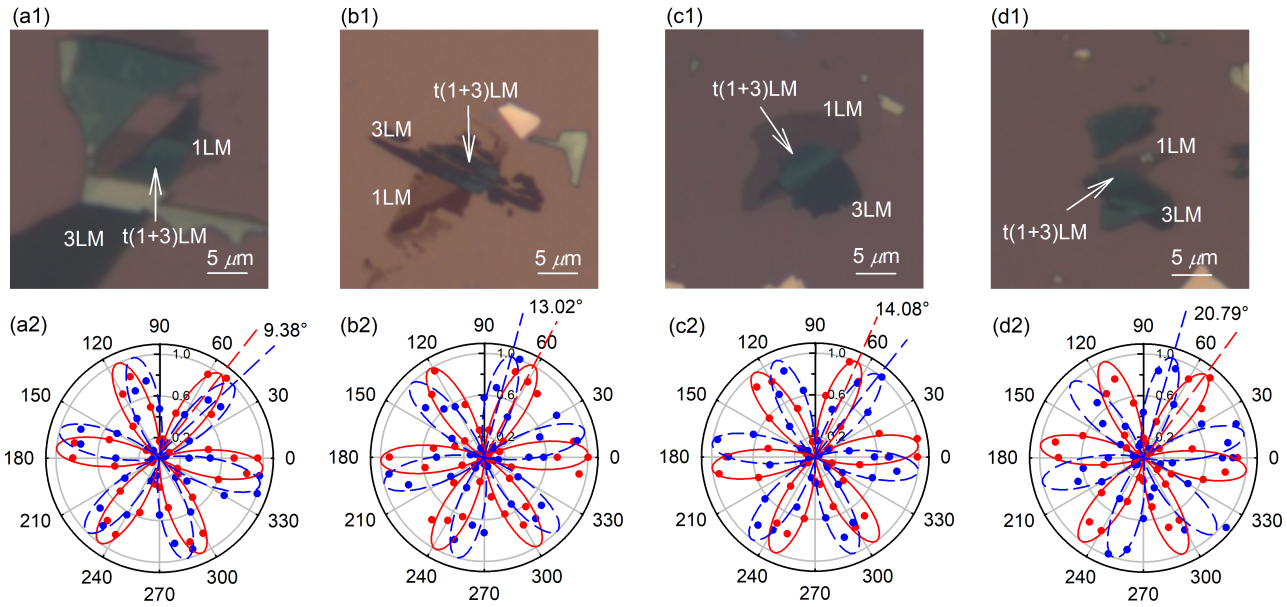

Fig. S5 (a1-d1) Optical image of four t(1+3)LM samples with different twist angles and (a2-d2) the crystallographic-orientation dependent SHG intensity of their 1LM (blue circle) and 3LM (red circle) constituents.

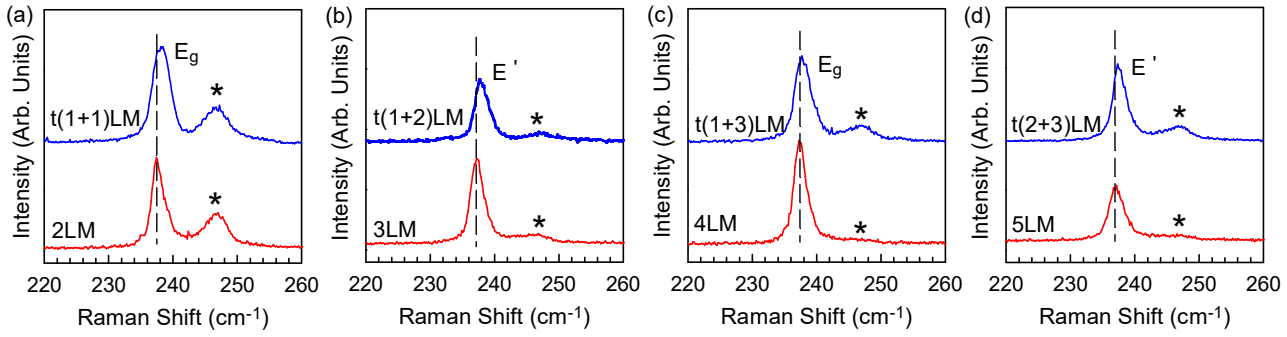

Fig. S6 Raman spectra of E'-like modes in $t(m+n)$LM and corresponding NLM ($N=m+n$): (a) t(1+1)LM and 2LM, (b) t(1+2)LM and 3LM, (c) t(1+3)LM and 4LM, (d) t(2+3)LM and 5LM. The combination modes, A_{1g}(M)+LA(M), are marked with asterisk. The vertical dashed lines are guides to eyes.