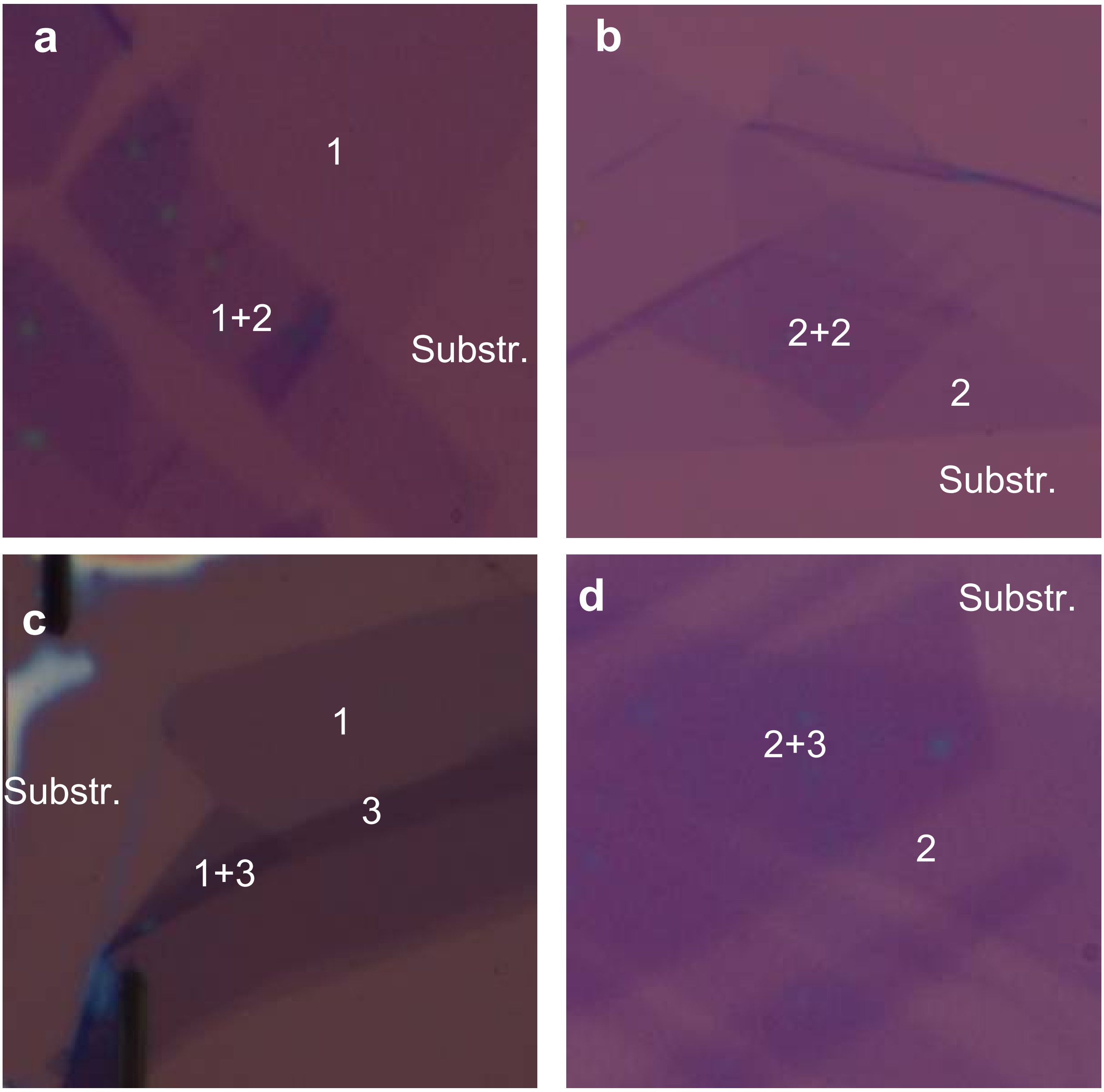
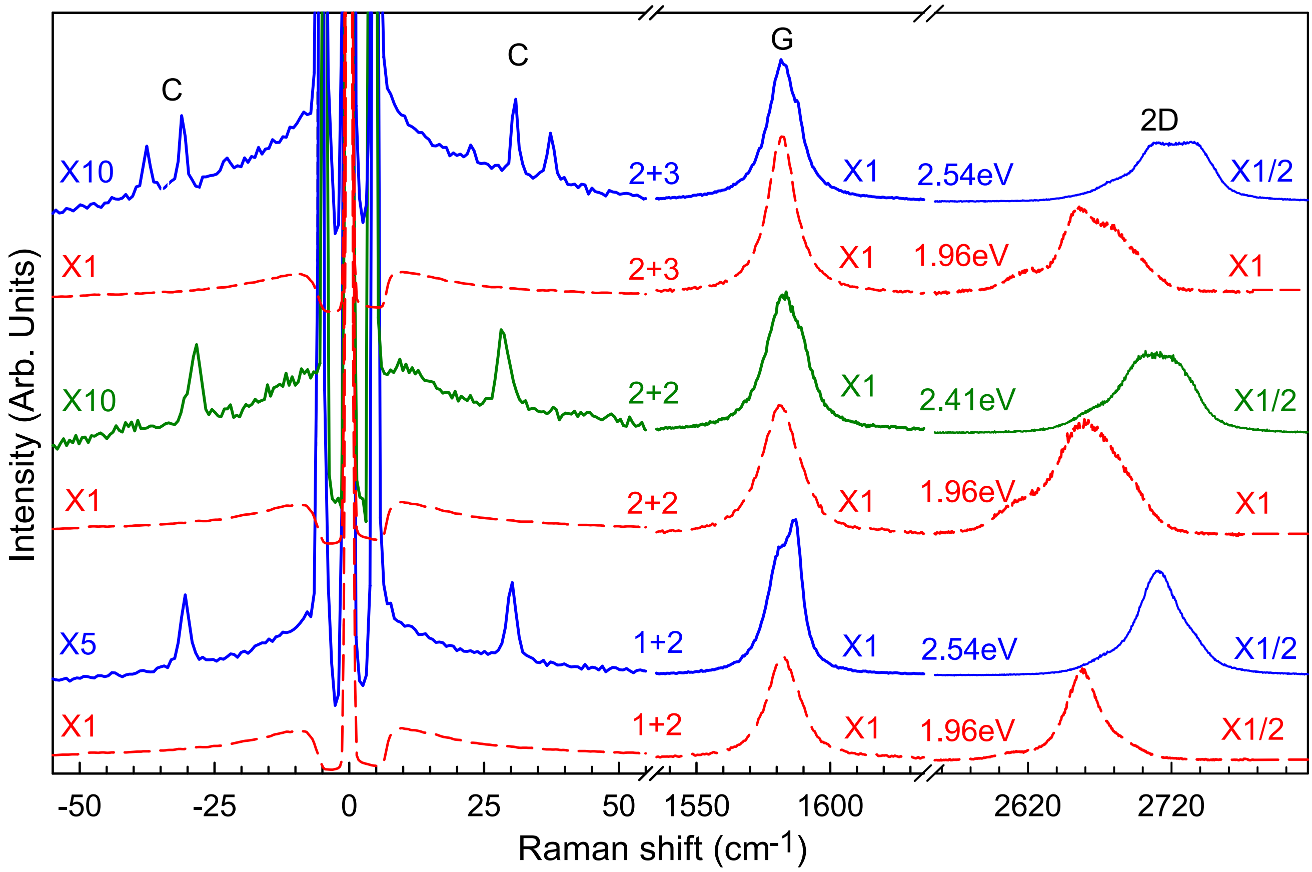
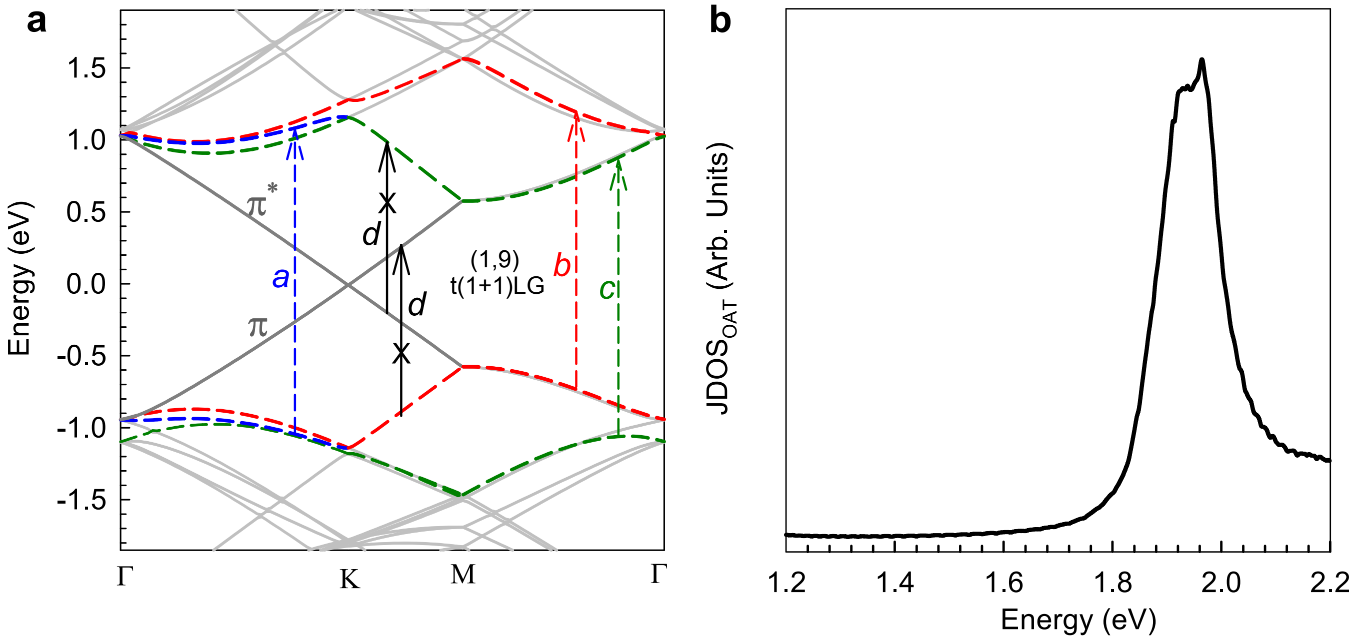
**Supplementary Figures**



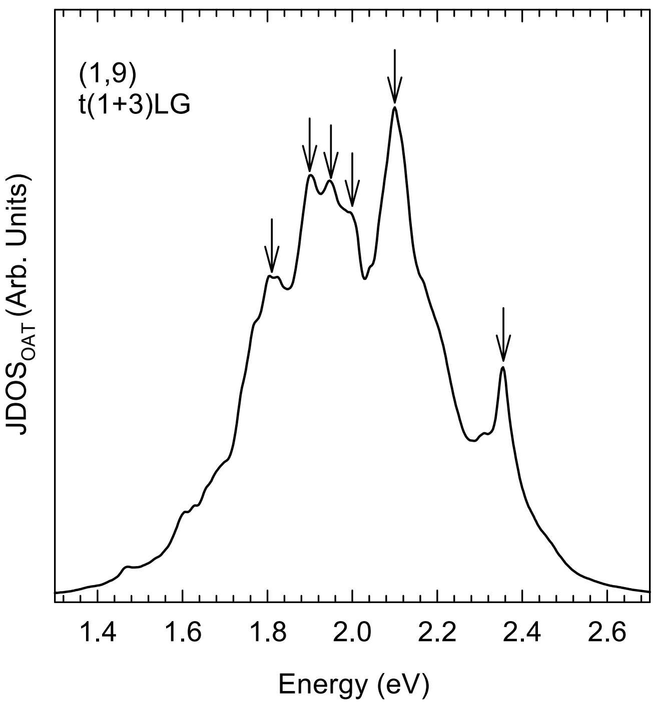

**Supplementary Figure 1 | Optical images of the t(m+n)LG samples.** (**a**) t(1+2)LG, (**b**) t(2+2)LG, (**c**) t(1+3)LG, and (**d**) t(2+3)LG. The numbers mark the number of layers in each region of the sample.




**Supplementary Figure 2 | Raman spectroscopy of t(m+n)LGs excited out-of-resonance and in-resonance.** The laser energy of out-of-resonance is 1.96 eV. The in-resonance energy of each t(m+n)LG are shown in the figure. No shear modes are observed for the t(1+2)LG, t(2+2)LG and t(2+3)LG at 1.96 eV (dashed lines). In contrast, the modes are clearly visible in the spectra measured within the resonant energy window (solid lines).




**Supplementary Figure 3 | JDOS$_{OAT}$ of t(1+1)LG.** (**a**) The band structure of (1,9) t(1+1)LG. Typical optically allowed transitions between the conduction and valence bands are shown with dashed arrows. The transitions between parallel bands connected by solid lines with solid crossed arrows are optically forbidden. (**b**) The corresponding the electronic joint density of states (JDOS) of all the optically allowed transitions (JDOS$_{OAT}$) of the band structure of (1,9) t(1+1)LG in (**a**).




**Supplementary Figure 4 | JDOS$_{OAT}$ of t(1+3)LG.** The six VHSs (at 1.81 eV, 1.90 eV, 1.95 eV, 1.99 eV, 2.10 eV and 2.36 eV) in the electronic joint density of states (JDOS) of all the optically allowed transitions (JDOS$_{OAT}$) are marked by arrows.

**Supplementary Note 1**

**Linear Chain Model for twisted multilayer graphene**

First, we consider a linear monatomic chain model for NLG. The equation of motion of this one-dimensional system can be written as[1]:

$$\begin{cases} m\ddot{U}_1 = -\alpha_0 U_1 + \alpha_0 U_2 \\ m\ddot{U}_2 = \alpha_0 U_1 - 2\alpha_0 U_2 + \alpha_0 U_3 \\ \vdots \\ m\ddot{U}_{N-1} = \alpha_0 U_{N-2} - 2\alpha_0 U_{N-1} + \alpha_0 U_N \\ m\ddot{U}_N = \alpha_0 U_{N-1} - \alpha_0 U_N \end{cases}, \tag{1}$$

where $m$ is the mass of carbon atom layer, $\alpha_0$ is the force constant between two layers, and $U_n$ is the shear displacement of the $n^{th}$ layer. The solution of the above equations of motion is obtained using the following substitution:

$$U_n = u_n \times e^{-i\omega t}, \tag{2}$$

Here, $u_n$ is the amplitude of displacement, and $\omega$ is the vibration frequency. After the substitution, the following equations are obtained:

$$\begin{cases} m\omega^2 u_1 = -\alpha_0 u_1 + \alpha_0 u_2 \\ m\omega^2 u_2 = \alpha_0 u_1 - 2\alpha_0 u_2 + \alpha_0 u_3 \\ \vdots \\ m\omega^2 u_{N-1} = \alpha_0 u_{N-2} - 2\alpha_0 u_{N-1} + \alpha_0 u_N \\ m\omega^2 u_N = \alpha_0 u_{N-1} - \alpha_0 u_N \end{cases}, \tag{3}$$

This can be expressed in a matrix form as:

$$m\omega^2 \mathbf{u} = \mathbf{D}\mathbf{u}, \tag{4}$$

This equation is equivalent to:

$$2\pi^2 c^2 \mu \omega^2 \mathbf{u} = \mathbf{D}\mathbf{u}, \tag{5}$$

where $\mu$=7.6×10$^{-27}$ kgÅ$^{-2}$ is the monolayer mass per unit area, $c = 3.0 \times 10^{10}$ cm s$^{-1}$ is the speed of light, $\mathbf{u}$ is the column vector of displacement, and $\mathbf{D}$ is the tridiagonal shear part of the force constant matrix. For example, the force constant matrix **D** for 5LG can be written as:

$$\mathbf{D} = \begin{bmatrix} -\alpha_0 & \alpha_0 & 0 & 0 & 0 \\ \alpha_0 & -2\alpha_0 & \alpha_0 & 0 & 0 \\ 0 & \alpha_0 & -2\alpha_0 & \alpha_0 & 0 \\ 0 & 0 & \alpha_0 & -2\alpha_0 & \alpha_0 \\ 0 & 0 & 0 & \alpha_0 & -\alpha_0 \end{bmatrix}, \tag{6}$$

There are N-1 non-zero frequencies $\omega_i$ and eigenvectors $\mathbf{u}_i$ that solve the equation. For each $\omega_i$, we can get the Eq (1) in the main text:

$$\omega_i^2 \mathbf{u}_i = \frac{1}{2\pi^2 c^2 \mu} \mathbf{D} \mathbf{u}_i, \tag{7}$$

For twisted multilayer graphene, for example t(2+3)LG, we denote the interlayer shear force constant between the graphene layers at the twisted interface $\alpha_t$. We also assume that the presence of the interface perturbs the force constant $\alpha_{0t}$ between the two layers adjacent to the interface. Then, the force constant matrix **D** for t(2+3)LG can be written as:

$$\mathbf{D} = \begin{bmatrix} -\alpha_{0t} & \alpha_{0t} & 0 & 0 & 0 \\ \alpha_{0t} & -\alpha_t - \alpha_{0t} & \alpha_t & 0 & 0 \\ 0 & \alpha_t & -\alpha_t - \alpha_{0t} & \alpha_{0t} & 0 \\ 0 & 0 & \alpha_{0t} & -\alpha_0 - \alpha_{0t} & \alpha_0 \\ 0 & 0 & 0 & \alpha_0 & -\alpha_0 \end{bmatrix}, \tag{8}$$

## References


[1] Born, M. & Huang, K. *Dynamical theory of crystal lattices*, vol. 188 (Clarendon Press Oxford, 1954).