Electronic Supplementary Information

Interlayer bond polarizability model for interlayer phonons in van der Waals heterostructures

Rui Mei[1,2], Miao-Ling Lin[1], Heng Wu[1,2], Lin-Shang Chen[1], Yan-Meng Shi[1], Zhong-Ming Wei[1] and Ping-Heng Tan[1,2*]

[1]State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China

[2]Center of Materials Science and Optoelectronics Engineering \& CAS Center of Excellence in Topological Quantum Computation, University of Chinese Academy of Sciences, Beijing 100049, China.

*Corresponding authors

E-mail: phtan@semi.ac.cn

Section I. Optical images of samples

The corresponding vdWHs were meticulously assembled layer by layer in specific orders, by employing a nanomanipulator. During sample preparation, a nail polish-coated polydimethylsiloxane (PDMS) block mounted on a glass slide was utilized to pick up and release 2DM[1,2]. Fig. S1 shows the optical images of the prepared samples, which exhibit high quality.

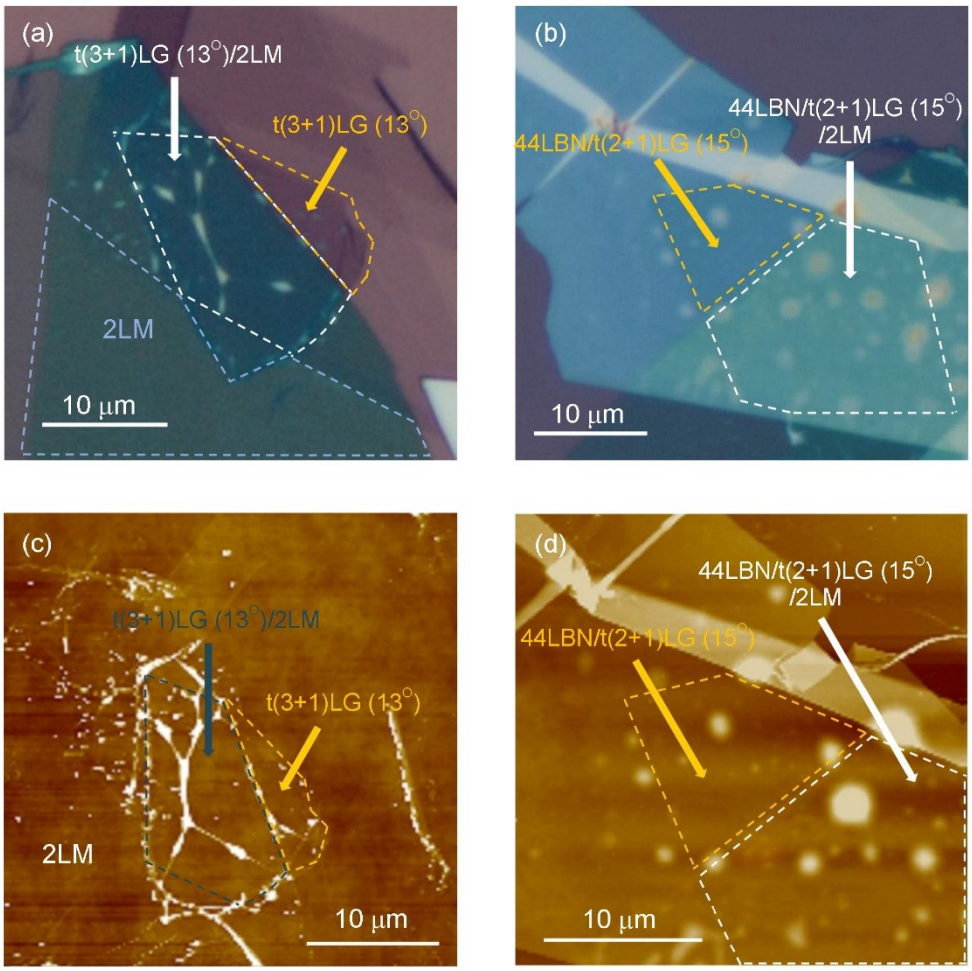

Fig. S1. Optical images and AFM images of samples. (a) Image of the sample which contains three regions of 2LM, t(3+1)LG 13° and t(3+1)LG 13°/2LM. (b) Optical image of the sample that includes 44L-hBN/t(2+1)LG 15° and 44L-hBN/t(2+1)LG 15 °/2LM. (c) AFM image of the sample containing 2LM, t(3+1)LG 13° and t(3+1)LG 13°/2LM regions. (d) AFM image of the sample depicted in (b).

Section II. Calculated frequencies of LBMs in vdWHs

For the LBMs in 2DMs or binary vdWHs, each layer vibrates rigidly and thus it can be simplified as a single rigid body with layer-breathing force constant interaction. Based on this feature, linear chain model (LCM) has been developed to predict the frequencies of LBMs in 2DMs and vdWHs, where only the nearest-neighbor interaction between adjacent rigid layers is considered[3,4]. However, the improved linear chain model

(2LCM) considering the interaction between second nearest-neighbor graphene layers is necessary to predict the LBMs in multilayer graphene (MLG), t(m+n)LG and its related vdWHs[5,6].

According to the 2LCM, the frequencies of the LBMs in tMLG-based vdWHs can be calculated by solving an N × N linear homogeneous equation[3–6]:

$$\omega_j^2 M u_j = \frac{1}{4\pi^2 c^2} D u_j \#(1)$$

where u_j is the interlayer displacements of $LB_{N,N-j}$ mode with frequency of ω_j, $\boldsymbol{M}$ is the diagonal mass matrix of the vdWH, $c = 3 \times 10^{10}$ cm/s and $\boldsymbol{D}$ is the LB force constant matrix. In our calculation, the LB force constant of MoS_2 is $\alpha^{\perp}(M) = 8.65 \times 10^{19}\, N \cdot m^{-3}$. The LB force constant of graphene is $\alpha^{\perp}(Gr) = 10.7 \times 10^{19}\, N \cdot m^{-3}$. The LB force constant between graphene and MoS_2 is $\alpha^{\perp}(Gr/M) = 5.68 \times 10^{19}\, N \cdot m^{-3}$. The LB force constant between graphene and hBN is $\alpha^{\perp}(Gr/hBN) = 7.83 \times 10^{19}\, N \cdot m^{-3}$. In addition, because the interaction between second-nearest-neighbor graphene layers is important, the second-nearest-neighbor LB force constant in tMLG constituent ($\beta^{\perp}(Gr) = 0.96 \times 10^{19}\, N \cdot m^{-3}$) should be considered. The calculated LBM frequencies of t(3+1)LG, t(2+1)LG, 44L-hBN/t(2+1)LG, t(3+1)LG/2LM, t(2+1)LG/2LM and 44L-hBN/ t(2+1)LG/2LM vdWHs are presented in Fig. S2. It is clear that the theoretical frequencies are in good agreement with the experimental ones.

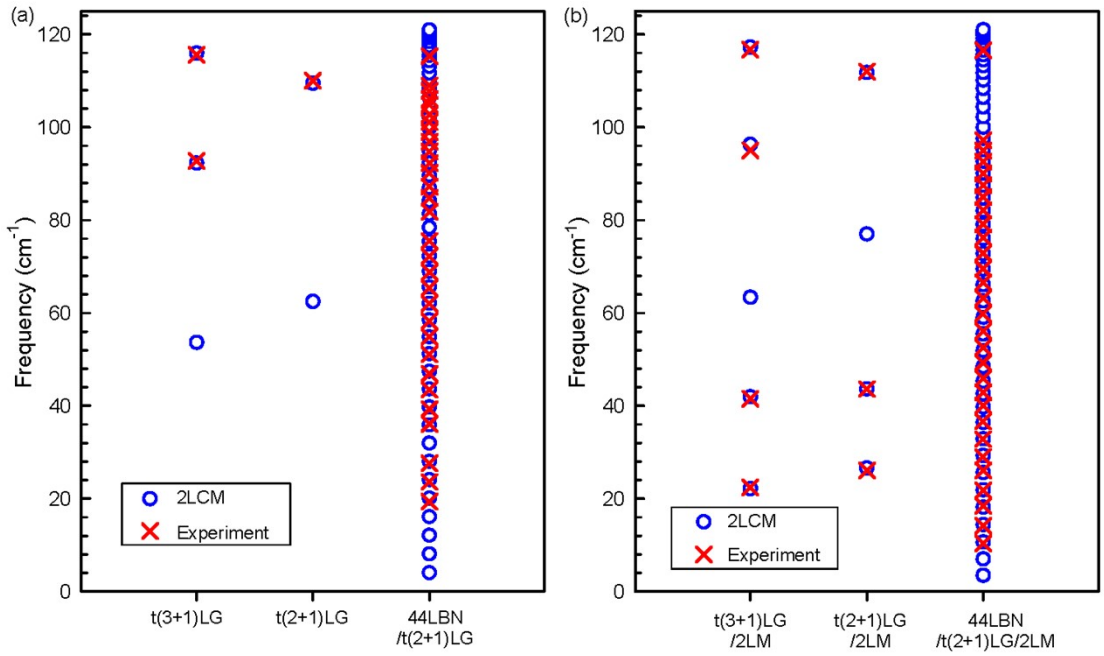

Fig. S2. (a) Theoretical (blue circles) and experimental (red crosses) LBMs frequencies of t(3+1)LG, t(2+1)LG and 44L-hBN/ t(2+1)LG. (b) Theoretical (blue circles) and experimental LBMs frequencies (red crosses) of t(3+1)LG/2LM, t(2+1)LG/2LM and 44L-hBN/ t(2+1)LG/2LM. The calculated results based on 2LCM are in good agreement with the experimental ones.

References

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