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Supplementary Materials for

Stacking symmetry governed second harmonic generation in graphene trilayers

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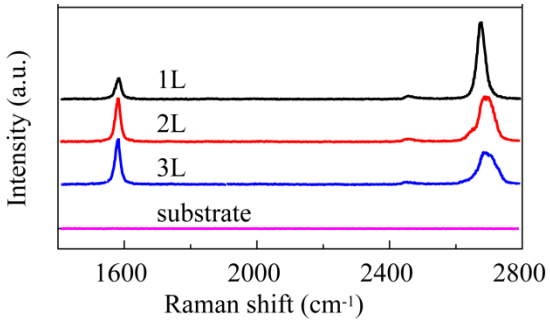

fig. S1. Raman spectra of the sample from a monolayer (1L), a bilayer (2L), and a trilayer (3L), along with the background from the bare substrate. The excitation wavelength was 532 nm. For the thinnest layer, the 2D peak (~2676 cm^{-1}) has a single Lorentzian line shape indicating a monolayer. For thicker layers, the 2D bands are broadened as composed of multiple peaks, and intensity of the G band (~1588 cm^{-1}) is roughly proportional to the number of layers.

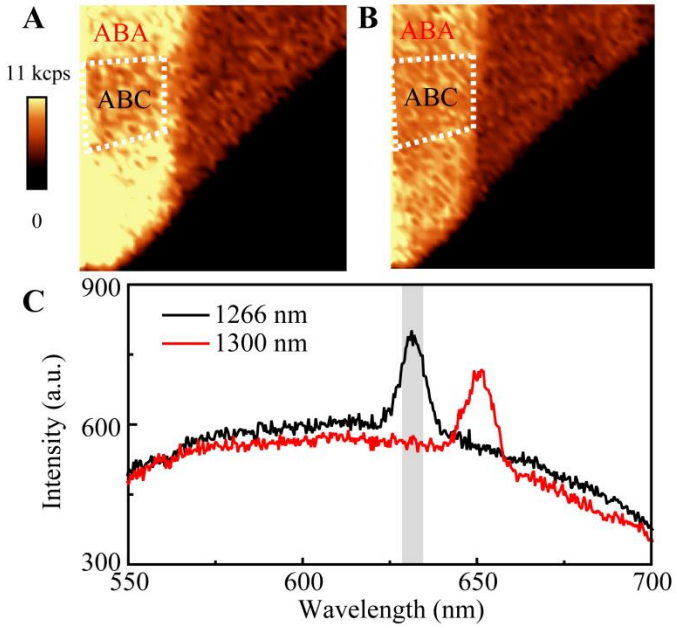

fig. S2. Comparison of nonlinear microscopy at different excitation wavelengths. (**A**) and (**B**) Nonlinear microscopy of the graphene trilayer in Fig. 4 with excitation wavelength of 1266 nm and 1300 nm, respectively. The dwell time at each pixel was 25 ms. (**C**) Corresponding nonlinear optical spectra from the ABA trilayer region. Both the excitation and signal beams were linearly polarized along the arm chair direction. The spectral bandpass range for nonlinear signal was centered at 633 nm and narrowed to 6 nm, which is grayed in the spectra.

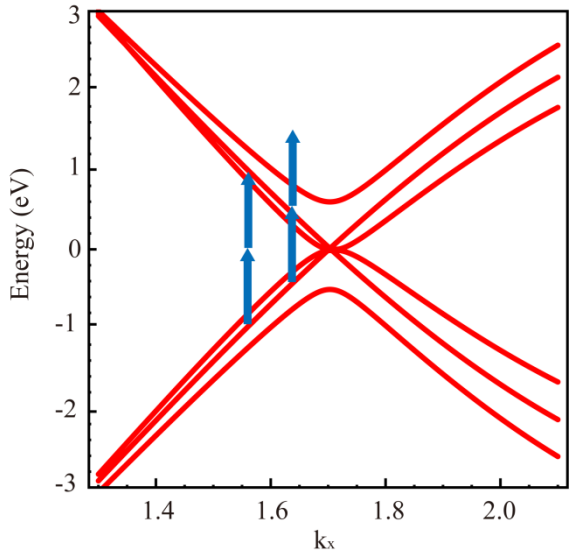

fig. S3. Schematic showing the resonant transitions in the SHG process. The one-photon or two-photon energy (blue arrows) could match with the interband transition in the band structure of ABA graphene trilayer (red curves). The resonance effects lead to the strong SHG we observed.

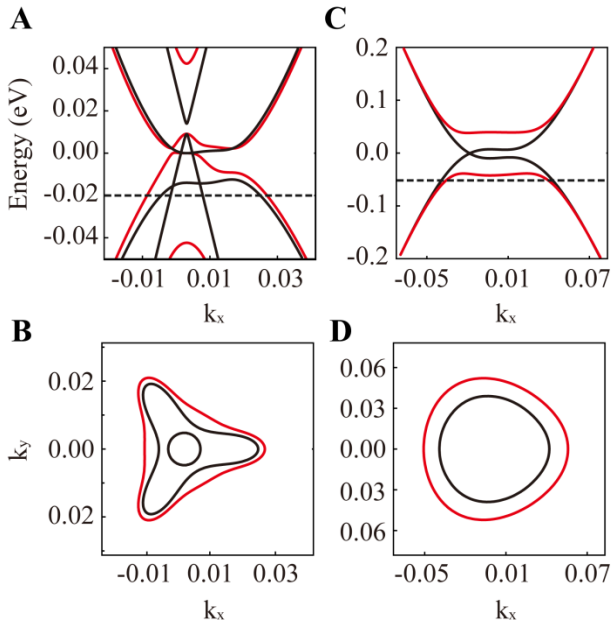

fig. S4. Stacking-dependent properties in ABA and ABC trilayers. (A) and **(B)** Calculated electronic band structure and contour of ABA trilayer, respectively. **(C)** and **(D)** Calculated electronic band structure and contour of ABC trilayer, respectively. The calculation followed the low-energy effective models (*36*, *37*). The black and red curves correspond to the band structures without (U_1 = 0 eV, U_2 = 0 eV, U_3 = 0 eV) and with (U_1 = 0.04 eV, U_2 = 0 eV, U_3 = -0.04 eV) a perpendicular electric field through the trilayer graphene, respectively. The contours in **(B)** and **(D)** are taken from the dotted line in **(A)** and **(C)** at the energy level of -0.02 eV and -0.05 eV, respectively. The effect of trigonal warping in ABA trilayer is stronger than that in ABC trilayer.