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Layer breathing vibrations in AB-stacking few layer graphene and twisted bilayer graphene

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Interlayer interactions in few layer graphene can create a set of shear modes and layer breathing modes (LBMs) that involve lateral and vertical displacement of individual layers, respectively. LBMs are of importance because they can facilitate interlayer current conduction and are sensitive to external perturbations, such as the presence of substrate or surface adsorbates. We investigated low-frequency fundamental layer breathing vibrations in AB-stacking few layer graphene and twisted bilayer graphene using Raman spectroscopy. In AB-stacking few layer graphene we observed the Raman peaks from phonons at the Brillouin zone center for the lowest-frequency branch LBM vibration. The mode frequency depends strongly on the number of graphene layers. Notably, the LBM Raman response is unobservable at room temperature, and it is turned on at higher temperature (>600 K) with a steep increase of Raman intensity. The observation suggests that the LBM vibration is strongly suppressed by molecules adsorbed on the graphene surface but is activated as desorption occurs at high temperature. In twisted bilayer graphene, the fundamental LBM is observed in a small range of twisting angle at which the intensity of the G Raman peak is strongly enhanced. The dependence of this mode's frequency and linewidth on the rotational angle can be explained by the double resonance Raman process (LBM phonon with nonzero wavevector) mediated by the twisted bilayer graphene lattice which lacks long-range translational symmetry. The angle dependence also reveals the strong impact of electronic band overlaps of the two rotated graphene layers.