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Observation of out-of-plane vibrations in few-layer graphene using combination and overtone Raman modes SUK HYUN KIM, CHUN HUNG LUI, LEANDRO MALARD, GABRIEL LANTZ, FRANÇOIS LAVERGE, Columbia University, RIICHIRO SAITO, Tohoku University, TONY HEINZ, Columbia University, HEINZ TEAM, SAITO TEAM — We have studied three distinct higher-order Raman features, appearing at ~ 1660 , 1730 and 1760 cm^{-1} , in graphene samples of 1-6 layers thickness and both Bernal and rhombohedral stacking. By detailed analysis of the measured dispersions of these lines using double-resonance theory, we have identified the features, respectively, as the LO+ZA, LO+ZO' combination modes and the 2ZO overtone mode. Here LO, ZA, and ZO, and ZO' denote, respectively, the in-plane longitudinal optical mode, the out-of-plane acoustic, optical and layer-breathing modes. All three of these Raman features are absent in single-layer graphene, which lacks the layer-breathing vibration and exhibits particularly high symmetry. The line shape of LOZO' mode shows a dramatic dependence on the stacking order of the layers and can serve as a means of identifying stacking order in few-layer graphene. In addition, the LOZO' mode allows us to access the properties of the low-energy layer-breathing (ZO') mode in few-layer graphene samples.

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