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Strong Rotational Angle Dependence of Raman Spectroscopy in Rotated Double-Layer Graphene KWANPYO KIM, SINISA COH, LIANG Z. TAN, WILLIAM REGAN, JONG MIN YUK, ERIC CHATTERJEE, M. F. CROMMIE, MARVIN L. COHEN, STEVEN G. LOUIE, A. ZETTL, Department of Physics, UC Berkeley — We perform a complementary Raman spectroscopy and transmission electron microscopy (TEM) study, as well as electronic-structure and Raman calculations, on suspended rotated double-layer graphene. Graphene Raman spectra show a strong dependence on the rotational angles between two stacked layers. For low-angle mis-orientations ($<\sim$ 10 degrees), double-layer graphene exhibits Raman signature closer to AB-stacked bilayer graphene. Double-layers with high rotational angles ($>\sim$ 15 degrees), on the other hand, display Raman spectra similar to monolayer graphene. Rotational angle dependent modifications of the electronic band structure in double-layer graphene can explain this trend and a G peak enhancement at certain middle angles. The computed electronic band structures and key features of the graphene Raman peaks including the blue shift, width and intensity of the 2D peaks agree well with experimental data.

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