Interest in two-dimensional (2D) materials has exploded in recent years following the isolation of graphene atomic layers from graphite crystal by the “scotch-tape” method in 2004. The numerous applications based on the exceptional electronic, optical, and mechanical properties of these materials are the real driving forces of research on 2D atomic layers. Raman spectroscopy is a convenient, noninvasive and powerful technique in probing the vibrational, optical, and electronic properties of materials. In this talk I will focus on our use of Raman techniques on 2D crystals to perform fundamental studies of interlayer phonons and charge ordering in these atomic layers. Our results reveal that the interface environment between the atomic layers and the substrate and the interlayer interactions within the atomic layers play an important role in modulating the properties of these 2D layered materials.