With an increasing need for wearable/stretchable devices in a variety of applications, there is strong demand for developing new materials that can preserve their intrinsic material properties intact in a harsh environment involving severe mechanical deformation. The limited mechanical flexibility inherent to covalently bonded inorganic thin film materials has sparked great interest in two-dimensional (2D) materials of extremely small thickness and high in-plane strain limits. Transition metal dichalcogenide (TMD) materials such as 2D molybdenum disulfide (2D MoS\textsubscript{2}) project unprecedented venues for 2D electronic/optoelectronic devices of unconventional form factors owing to their tunable bandgap energy and semiconducting transport coupled with structural robustness. Although it has been theoretically predicated that 2D MoS\textsubscript{2} layers can present the high in-plane strain limit of ~30\% owing to their near atomic thickness, chemically-grown 2D MoS\textsubscript{2} layers generally exhibit much limited stretchability - typically less than ~7\% strain. Such drawbacks severely limit the versatility of 2D MoS\textsubscript{2} layers for applications requiring a balanced combination of superior electrical/optical properties and large mechanical tolerance. In this talk, I will discuss about a viable strategy to three-dimensionally architect 2D MoS\textsubscript{2} layers into a tailored geometry which can ensure the desired high mechanical flexibility/stretchability preserving their intrinsic electrical/optical properties. I developed a “water-assisted 2D layer transfer” process by which I integrated periodically-corrugated 2D MoS\textsubscript{2} layers on elastomer polymer substrates precisely maintaining their tailored dimensional features. By carrying out extensive and systematic material property characterizations, I demonstrated the highly tunable electrical, optical, and surface properties of the periodically-corrugated 2D MoS\textsubscript{2} layers under varying tensile stretch. Particularly, these 3D-structured 2D MoS\textsubscript{2} layers retain excellent electrical conductivity even under stretch up to 50\% while their optical absorbance and surface wettability are simultaneously modulated. Lastly, I will discuss about the potential of these novel 2D materials for a broad range of unconventional applications such as wearable E-skin sensors.