The ability to engineer quantum electronic states in low-dimensional material systems has created opportunities for new device concepts and their integration into next generation technologies. In particular, atomically thin layered materials and their heterostructures provide an excellent material platform on which to build quantum electronic, optoelectronic, and thermoelectric devices. The primary goal of this Vannevar Bush Faculty Fellowship (VBFF) proposal is to design and engineer topological electronic structures using hetero-interfaces of atomically thin layered materials to enable the development of novel quantum electronic and optoelectronic devices. The recent rise of layered van der Waals (vdW) materials allows for a new type of layered heterostructures with strong quantum confinement. The atomically sharp interfaces of vdW heterostructures provide ample opportunity to discover topological quantum phenomena occurring at the interface between dissimilar atomic layers. Often, low dimensional interfacial electronic systems with strong spin-orbit coupling (SOC) can introduce a unique spin texture into the electronic structures, which can in turn exhibit topological electronic and optoelectronic properties. These robust quantum effects can be further utilized for novel device applications. This research proposal is based on our recently developed capability to create vdW interfaces with atomic precision. Building on our existing techniques, we will establish a fabrication procedure for topologically unique quantum heterostructures. Notably, we can achieve atomically precise sample manipulation and characterization in an entirely controlled environment by utilizing a novel device processing instrument. This unique capability allows us to consistently produce vdW heterostructures of exceptional quality. These heterostructures will be used to study correlated interfacial states, where quantum functionalities can be engineered by vdW interfacial proximity to exhibit topologically unique electronic characteristics. We will choose two electrically dissimilar vdW materials such as layered superconductors, P-type and N-type SOC semiconducting materials, and topological insulators to create novel interfaces. Using combined transport, local probe, and optical spectroscopic experimental techniques, we will explore the topological electronic and optoelectronic properties of the constructed interfacial states. Our experimental approach will be guided by collaboration with theorists to establish the rationale for the materials selection to realize the targeted functionality. Specifically, we will investigate (1) the development of topological superconductivity by proximitizing chiral topological states based on ferromagnetic/topological insulators or quantum Hall systems; (2) the interlayer exciton condensation in PN atomically thin semiconductor heterostructures; and (3) the quantum thermoelectric effect between PN topological insulator junctions. These systems offer immense promise for the manifestation of emergent physical phenomena, leading to groundbreaking scientific discoveries. Ultimately, our understanding and the technology developed in this project will enable the pursuit of innovative device applications, including efficient quantum electronics, optoelectronics, and thermoelectrics. Such novel device components will be important to the technological needs of the DoD, including data communication, computing, signal processing, and energy conversion. The multidisciplinary nature of the proposed research will also provide training to the next generation of technical experts who will contribute to the DoD’s mission.