One of the key advances in nanoscience and nanotechnology has been our increasing ability to reach the limits of atomically precise structures. By having developed the “eyes” to see, to record spectra, and to measure function at the nanoscale, we have been able to fabricate structures with precision. The physical, electronic, mechanical, and chemical connections that materials make to one another and to the outside world are critical. Just as the properties and applications of conventional semiconductor devices depend on these contacts, so do nanomaterials, many nanoscale measurements, and devices of the future.