ABSTRACT: Our first atomic-resolution measurements of isolated and dilute interacting atoms and molecules on surfaces with scanning tunneling microscopy and spectroscopy revealed long-range, anisotropic, energy-dependent perturbations on electronic structure. Spectroscopic imaging enables energy- and spatially resolved measurements of electronic structure, but we have not yet been able to invert these perturbations directly into adsorbate potentials. Thus far, we have used measurements of adsorbate structures and dynamics to extract these potentials quantitatively with unprecedented precision. These long-range effects have important consequences in catalysis and supramolecular assembly. Next, we will attempt to design these interactions to advantage to control the placement and transformation of atoms and molecules on surfaces.