This presentation explores the development of NMR and modeling methods designed to provide crystal structures for materials that are not suitable for conventional diffraction methods. One area of focus is a highly promising non-diffraction alternative known as crystal structure prediction (CSP) - the accurate computational prediction of solid-state structure solely from the knowledge of atomic connectivity. The promise of CSP arises from its complete independence from diffraction data, eliminating difficulties associated with diffraction (e.g. poor x-ray diffraction from lighter elements). Despite over three decades of intense research, however, theoretical prediction of crystal structure has remained an elusive goal. Recent work in our laboratory demonstrates that when solid-state NMR data from powders are included in the CSP process, the correct structure can consistently be identified from among dozens of candidates. Presently, most CSP structures are of low quality and even when a correct structure is identified, a refinement step is needed to “clean up” the structure. Thus, a second area of emphasis is the theoretical refinement of CSP structures using DFT methods that include lattice effects. Solid-state NMR chemical shift tensors (\(^{13}\)C and \(^{15}\)N) are remarkably sensitive to these refinements and are used in our lab to monitor refinements. This NMR work suggests that such refinements have the potential to create ultra-high resolution structures from CSP and more conventional diffraction data that rival the accuracy of single crystal neutron diffraction coordinates.