Over the past two decades, terahertz time-domain spectroscopy (THz-TDS) has become a valuable technique for the characterization of solid samples, primarily due to its sensitivity to bulk molecular packing arrangements, such as in complex pharmaceutical molecular crystals. However, in recent years the role that specific terahertz vibrations (> ~330 cm⁻¹ or ~10 THz) play in a number of important physical phenomena has become increasingly apparent, with numerous studies highlighting how terahertz motions are directly responsible for the proper functioning of materials, ranging from enzymatic catalysis to solid-state phase transformations. Such dynamics are intimately linked to the molecular structures, condensed phase geometries, and even individual atomic details (e.g. electronic configuration), and therefore understanding material properties is intrinsically linked to understanding the associated low-frequency motions. In this work, the nature of terahertz-frequency motions in molecular solids will be used to determine a number of physical characteristics of biomolecular and pharmaceutical materials. Initially, a review of THz-TDS will be presented, with an example of how the prototypical terahertz vibrations in a biological polypeptide polymer can be used as an effective method of determining mechanical properties, such as bulk elasticity. Subsequently, the terahertz motions in disordered (amorphous) active pharmaceutical ingredients (APIs) will be shown to be a powerful indicator for predicting the stability of these materials. Moreover, the terahertz dynamics can be exploited to induce polymorph-specific crystallization by exciting the sample at certain distinctive frequencies corresponding to particular motion-types. Finally, the role that terahertz vibrations play in phase transition mechanisms of molecular crystals will be presented, and the conclusive link between the two will be highlighted. Collectively, these findings provide a compelling demonstration that terahertz radiation can be used to direct the properties and structures of condensed phase materials through their low-frequency vibrations, ultimately enabling new approaches for designing advanced materials.