The quantum nature of materials spans microscopic to macroscopic scales. This enables a wide array of physical properties that facilitate applications such as energy relevant technologies. The exotic properties arise from intertwined couplings of symmetry, topology, dimensionality and strong correlations, and are sensitive to external stimuli. We can use this sensitivity to unravel the complex interplay between the degrees of freedom by perturbing the interaction parameters and observing the corresponding responses. Pressure provides a clean and effective tuning parameter, but introduces challenges to successfully performing in situ measurements, especially at low temperatures. In this talk, I will discuss how we employ complementary optical techniques to probe the quantum physics of the strongly spin-orbit-coupled Mott insulator and Weyl semimetal under extreme conditions. Taken together, these experiments open new windows into quantum materials by providing multiple probes for accessing and studying novel phases.