At last year’s Symposium, we reported the rapid vapor deposition of millimeters thick parahydrogen (pH2) solids of remarkable optical clarity. In this paper we present our progress towards understanding the microscopic structure of these samples, as well as a potpourri of spectra of trapped molecular species illustrating some of the advantages of performing matrix isolation spectroscopy (MIS) in these samples. Infrared (IR) and Raman spectra of pure pH2 samples show a very low orthohydrogen and vacancy content, and a mixed hcp/fcc polycrystalline structure for as-deposited samples, which converts to hcp upon annealing . The increased optical path lengths offer significant improvements in spectroscopic data quality, and reveal novel dopant-induced IR absorptions of the pH2 matrix host itself. Thus, while traditional MIS studies in rare gas hosts can only probe the influence of the matrix environment on the spectrum of the dopant ”solute,” in pH2 the response of the host ”solvent” is directly observable as well. This complementary information may prove key to identifying conclusively the microscopic structures of dopant trapping sites.