SPECTRAL INHOMOGENEITY AS A DISCRETE, COUNTABLE ENTITY – THE ROLE OF RESIDUAL ORTHO-HYDROGEN MOLECULES ON THE ROVIBRATIONAL SPECTRA OF DOPANTS IN PARAHYDROGEN SOLIDS

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We report high resolution (0.005 cm^{-1}) infrared (IR) absorption spectra of doped millimeters-thick parahydrogen (pH_2) solids with intentionally elevated residual orthohydrogen (oH_2) concentrations. In addition to the weakly-hindered rotor spectrum of isolated H_2O molecules, $H_2O/oH_2/pH_2$ samples exhibit a series of sharp (0.03 cm^{-1} FWHM) absorption peaks in the 1593 to 1597 cm^{-1} region which we show are due to H_2O-oH_2 clusters. We have also identified CO_2-oH_2 clusters as the source of the very sharp (0.007 cm^{-1} FWHM) fine structure in IR spectra of $CO_2/oH_2/pH_2$ solids.[a] The gradual evolution of the CO_2-oH_2 features with increasing oH_2 concentration raises the interesting notion of spectral inhomogeneity as a discrete, countable entity.

[a] S. Tam and M.E. Fajardo, Low Temp. Phys. v26, p653 (1998).