Proton transfer reactions are essential in many biological and chemical processes, but are notoriously difficult to study directly due to strong anharmonic effects and ultrafast vibrational relaxation dynamics. Here, we present ultrafast transient absorption (TA) and two-dimensional infrared (2DIR) spectroscopies to characterize the intramolecular H-bonding interactions and proton transfer dynamics in a series of acetylacetone derivatives and their deuterated isotopologues. Strong 2DIR cross peaks are observed in the OH/OD bend region, indicating significant coupling between the OH/OD bends and other fingerprint modes. The OH/OD stretch regions display broad, homogeneous ground-state bleach signals that decay within 100 fs to intramolecular modes, resulting in long-lived hot-ground state signatures. Interestingly, polarization-dependent measurements reveal anisotropic signals that persist beyond the vibrational lifetimes with large isotopic differences. In general, the OH stretch anisotropies decay within about 200 fs, equivalent to one period of the intramolecular O-O stretch H-bond soft mode. The OD stretch anisotropies decay within 1-2 ps and correlate with the H-bond strength. The observed anisotropy timescales are interpreted in terms of the delocalization and transfer timescales of the proton/deuteron upon vibrational excitation.