Purpose: The "Magic Angle Effect" observed in cartilage, tendon and other collagen rich tissues of the extracellular matrix have largely unused potential for evaluation of musculo-skeletal disease processes. These studies seek to improve understanding of the molecular basis for magic angle phenomena based on collagen hydration. We hypothesize that improved understanding will provide a conceptual framework and extend the ability of musculo-skeletal radiologists to design protocols using MRI orientational contrast for better accuracy and specificity. Method and Materials: These studies use Differential Scanning Calorimetry (DSC) to directly measure the enthalpy and entropy of water bound to rat tail collagen/tendon at different hydration levels. Water vapor sorption and recovery rehydration rates of rat tail collagen/tendon at 22 °C were also measured. Measured bound water fractions are compared to the theoretical values (h=0.263 g/g) predicted from the molecular structure of collagen. Bound water hydration measured with both methods agreed with the molecular prediction of water bound to the protein back bone. Statistical analyses performed using Graphpad Prism. Results: The water vapor sorption and DSC studies of collagen/tendon at 22 °C show that both equilibrium hydration and enthalpy are linear functions of relative humidity up to critical hydration of the protein backbone h = 0.26 g/g. The water bridge hydration hypothesis identifies three hydration water fractions in direct contact with the protein that differ in motional/orientational properties from bulk water. Comparison to T1 and T2 relaxation rate and orientational studies shows the relaxation rate of tendon is determined by fast exchange of water between these motional and orientational restricted water fractions. Conclusion: This work offers evidence for the important role of protein main chain hydration in determining MRI contrast due to dielectric binding of polar water molecules interacting with partial electric charges separated on the backbone by steric restrictions of the collagen molecule.