

Title: Do Hydration Dynamics Follow the Structural Perturbation during Thermal Denaturation of a Protein: A Terahertz Absorption Study

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Abstract: We investigate the thermal denaturation of human serum albumin and the associated solvation using terahertz (THz) spectroscopy in aqueous buffer solution. Far- and near-ultraviolet circular dichroism spectroscopy reveal that the protein undergoes a native (N) to extended (E) state transition at temperature ≤ 55 degrees C with a marginal change in the secondary and tertiary structure. At 70 degrees C, the protein transforms into an unfolded (U) state with significant irreversible disruption of its structures. We measure the concentration- and temperature-dependent THz absorption coefficient (α) of the protein solution using a p-Ge THz difference spectrometer (2.1-2.8 THz frequency range), thereby probing the collective protein-water network dynamics. When the solvated protein is heated up to 55 degrees C and cooled down again, a reversible change in THz absorption is observed. When increasing the temperature up to 70 degrees C, we find a dramatic irreversible change of THz absorption. The increase in THz absorption compared to bulk water is attributed to a blue shift in the spectrum of the solvated protein compared to bulk water. This is supported by measurements of THz absorption coefficients using THz time-domain spectroscopy (0.1-1.2 THz frequency range). We also use picosecond-resolved fluorescence spectroscopy of the tryptophan 214 moiety of human serum albumin. All experimental observations can be explained by a change in the hydration dynamics of the solvated protein due to the additional exposure of hydrophobic residues upon unfolding.