

# Dynamics of a Globular Protein and Its Hydration Water Studied by Neutron Scattering and MD Simulations

Chansoo Kim<sup>1</sup>, Xiang-qiang Chu<sup>2</sup>, Marco Lagi<sup>3</sup>, Sow-Hsin Chen<sup>4</sup>, Kwang-Ryeol Lee<sup>1</sup>

<sup>1</sup>Computational Science Center, Korea Institute of Science and Technology, <sup>2</sup>Oak Ridge National Laboratory, <sup>3</sup>New England Complex Systems Institute, <sup>4</sup>Applied Physics, Dept't of Nuclear Science, Massachusetts Institute of Technology

A series of Quasi-Elastic Neutron Scattering (QENS) experiments helps us to understand the single-particle (hydrogen atom) dynamics of a globular protein and its hydration water and strong coupling between them. We also performed Molecular Dynamics (MD) simulations on a realistic model of the hydrated hen-egg Lysozyme powder having two proteins in the periodic box.

We found the existence of a Fragile-to-Strong dynamic Crossover (FSC) phenomenon in hydration water around a protein occurring at  $T_L=225\pm 5\text{K}$  by analyzing Intermediate Scattering Function (ISF). On lowering of the temperature toward FSC, the structure of hydration water makes a transition from predominantly the High Density Liquid (HDL) form, a more fluid state, to predominantly the Low Density Liquid (LDL) form, a less fluid state, derived from the existence of a liquid-liquid critical point at an elevated pressure.

We showed experimentally and confirmed theoretically that this sudden switch in the mobility of the hydration water around a protein triggers the dynamic transition (so-called glass transition) of the protein, at a temperature  $T_D=220\text{K}$ . Mean Square Displacement (MSD) is the important factor to show that the FSC is the key to the strong coupling between a protein and its hydration water by suggesting  $T_L \approx T_D$ .

MD simulations with TIP4P force field for water were performed to understand hydration level dependency of the FSC temperature. We added water molecules to increase hydration level of the protein hydration water, from 0.30, 0.45, 0.60 and 1.00 (1.00 is the bulk water). These confirm the existence of the FSC and the hydration level dependence of the FSC temperature: FSC temperature is decreased upon increasing hydration level.

We compared the hydration water around Lysozyme, B-DNA and RNA. Similarity among those suggests that the FSC and this coupling be universal for globular proteins, biopolymers.

**Keywords:** water, super cooled water, protein hydration water, lysozyme, glass transition temperature, molecular dynamics, neutron scattering