***Supplementary information:***

# *Nanosecond relaxation dynamics of hydrated proteins: Water versus protein contributions*

S. Khodadadi[1*], J. E. Curtis[1], A. P. Sokolov[2]

[1]NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899
[2]Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831; Department of Chemistry, University of Tennessee, Knoxville, Tennessee 37996

* The corresponding author: sheila.khodadadi@nist.gov

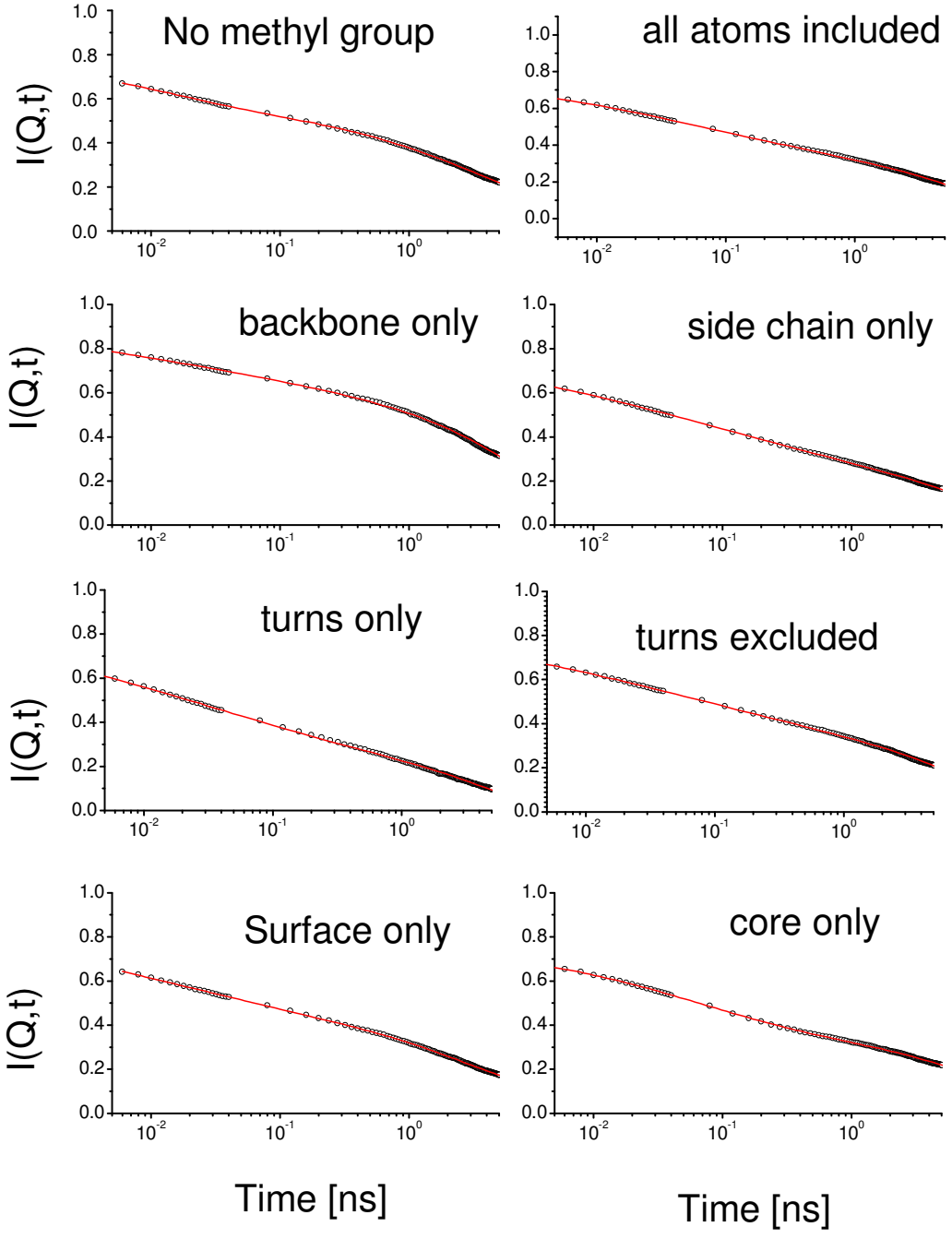


Fig S1. Calculated I (Q,t) of protein atoms at Q=2 $Å^{-1}$, symbols. The data have been fitted by $y_0=y_0+A_{main} \exp(-x/\tau_{main})^{\alpha}{}_{main}+A_{slow} \exp(-x/\tau_{slow})^{\alpha}{}_{slow}$, shown in lines. $R^2 > 0.97$.

Table S1- Fit parameters for calculated I (Q,t) of protein atoms with $y_0=y_0+A_{main} \exp(-x/\tau_{main})^{\alpha}{}_{main}+A_{slow} \exp(-x/\tau_{slow})^{\alpha}{}_{slow}$ when all the atoms are included.

| Q | $y_0$ | $A_{main}$ | $\tau_{main}$ (ns) | $\alpha_{main}$ | $A_{slow}$ | $\tau_{slow}$ (ns) | $\alpha_{slow}$ |
|---|---|---|---|---|---|---|---|
| 1 | 0.00±0.44 | 0.18±0.09 | 0.04±0.03 | 0.38±0.05 | 0.78±0.52 | 29.85±44.04 | 0.54±0.21 |
| 1.5 | 0.00±0.72 | 0.36±0.05 | 0.07±0.02 | 0.42±0.04 | 0.52±0.11 | 12.31±2.65 | 0.77±0.14 |
| 2 | 0.00±0.05 | 0.45±0.08 | 0.04±0.02 | 0.37±0.04 | 0.37±0.11 | 8.35±1.20 | 0.76±0.20 |
| 2.5 | 0.00±0.03 | 0.43±0.09 | 0.04±0.02 | 0.37±0.05 | 0.25±0.10 | 6.25±0.83 | 0.76±0.26 |
| 3 | 0.00±0.01 | 0.64±0.13 | 0.01±0.00 | 0.23±0.45 | 0.15±0.07 | 5.54±0.60 | 0.75±0.21 |

Table S2- Fit parameters for calculated I (Q,t) of protein atoms with $y_0=y_0+A_{main} \exp(-x/\tau_{main})^{\alpha}{}_{main}+A_{slow} \exp(-x/\tau_{slow})^{\alpha}{}_{slow}$ when methyl groups are excluded.

| Q | $y_0$ | $A_{main}$ | $\tau_{main}$ (ns) | $\alpha_{main}$ | $A_{slow}$ | $\tau_{slow}$ (ns) | $\alpha_{slow}$ |
|---|---|---|---|---|---|---|---|
| 1 | 0.01±0.36 | 0.15±0.12 | 0.02±0.02 | 0.33±0.15 | 0.81±0.44 | 26.23±29.34 | 0.56±0.18 |
| 1.5 | 0.00±0.06 | 0.21±0.03 | 0.01±0.00 | 0.40±0.04 | 0.71±0.08 | 10.08±2.24 | 0.53±0.05 |
| 2 | 0.01±0.01 | 0.18±0.03 | 0.01±0.00 | 0.61±0.08 | 0.59±0.02 | 4.78±0.27 | 0.48±0.02 |
| 2.5 | 0.00±0.00 | 0.29±0.02 | 0.01±0.00 | 0.46±0.03 | 0.43±0.01 | 3.58±0.11 | 0.54±0.01 |
| 3 | 0.00±0.00 | 0.36±0.03 | 0.01±0.00 | 0.39±0.03 | 0.30±0.01 | 2.90±0.15 | 0.58±0.02 |

Table S3- Fit parameters for calculated I (Q,t) of protein backbone atoms with $y_0=y_0+A_{main} \exp(-x/\tau_{main})^{\alpha}{}_{main}+A_{slow} \exp(-x/\tau_{slow})^{\alpha}{}_{slow}$.

| Q | $y_0$ | $A_{main}$ | $\tau_{main}$ (ns) | $\alpha_{main}$ | $A_{slow}$ | $\tau_{slow}$ (ns) | $\alpha_{slow}$ |
|---|---|---|---|---|---|---|---|
| 1 | 0.00±0.20 | 0.17±0.37 | 0.01±0.02 | 0.19±0.39 | 0.45±0.18 | 10.66±1.97 | 0.83±0.17 |
| 1.5 | 0.06±0.14 | 0.23±0.17 | 0.02±0.02 | 0.29±0.18 | 0.70±0.23 | 12.28±3.74 | 0.72±0.18 |
| 2 | 0.00±0.07 | 0.52±0.94 | 0.02±0.06 | 0.21±0.35 | 0.53±0.49 | 9.369±1.72 | 0.80±0.34 |
| 2.5 | 0.00±0.04 | 0.52±0.37 | 0.02±0.01 | 0.28±0.18 | 0.42±0.20 | 6.45±0.81 | 0.83±0.25 |
| 3 | 0.00±0.01 | 0.64±0.37 | 0.02±0.01 | 0.26±0.14 | 0.26±0.15 | 5.55±0.54 | 0.93±0.27 |

Table S4- Fit parameters for calculated I (Q,t) of protein side chain atoms with $y_0=y_0+A_{main} \exp(-x/\tau_{main})^{\alpha}{}_{main}+A_{slow} \exp(-x/\tau_{slow})^{\alpha}{}_{slow}$.

| Q | $y_0$ | $A_{main}$ | $\tau_{main}$ (ns) | $\alpha_{main}$ | $A_{slow}$ | $\tau_{slow}$ (ns) | $\alpha_{slow}$ |
|---|---|---|---|---|---|---|---|
| 1 | 0.05±0.47 | 0.44±0.22 | 0.49±0.98 | 0.31±0.05 | 0.48±0.67 | 28.42±38.75 | 0.95±0.54 |
| 1.5 | 0.00±0.16 | 0.61±0.19 | 0.13±0.15 | 0.28±0.05 | 0.35±0.30 | 16.45±10.44 | 0.88±0.40 |
| 2 | 0.00±0.06 | 0.59±0.11 | 0.05±0.03 | 0.30±0.04 | 0.27±0.15 | 10.10±1.65 | 0.80±0.30 |
| 2.5 | 0.00±0.00 | 0.52±0.05 | 0.02±0.00 | 0.32±0.03 | 0.23±0.02 | 5.89±0.75 | 0.69±0.05 |
| 3 | 0.00±0.00 | 0.44±0.36 | 0.01±0.00 | 0.33±0.02 | 0.18±0.01 | 3.94±0.48 | 0.62±0.04 |

Table S5- Fit parameters for calculated I (Q,t) of protein atoms of turns with $y_0=y_0+A_{main}$ exp (-x/ $\tau_{main}$) $^{\alpha}{}_{main}+A_{slow}$ exp (-x/ $\tau_{slow}$) $^{\alpha}{}_{slow}$.

| Q | $y_0$ | $A_{main}$ | $\tau_{main}$ (ns) | $\alpha_{main}$ | $A_{slow}$ | $\tau_{slow}$ (ns) | $\alpha_{slow}$ |
|---|---|---|---|---|---|---|---|
| 1 | 0.04±0.15 | 0.38±0.30 | 0.09±0.26 | 0.30±0.12 | 0.59±0.40 | 13.21±3.52 | 0.76±0.32 |
| 1.5 | 0.00±0.00 | 0.59±0.1 | 0.06±0.02 | 0.30±0.04 | 0.37±0.06 | 7.74±0.65 | 0.88±0.07 |
| 2 | 0.00±0.09 | 0.69±0.09 | 0.03±0.00 | 0.29±0.04 | 0.23±0.04 | 5.13±0.41 | 0.96±0.10 |
| 2.5 | 0.00±0.00 | 0.76±0.09 | 0.01±0.00 | 0.26±0.03 | 0.15±0.03 | 3.54±0.30 | 0.86±0.07 |
| 3 | 0.00±0.00 | 0.69±0.10 | 0.01±0.00 | 0.26±0.04 | 0.08±0.02 | 3.21±0.39 | 1.00±0.16 |

Table S6- Fit parameters for calculated I (Q,t) of protein atoms with $y_0=y_0+A_{main}$ exp (-x/ $\tau_{main}$) $^{\alpha}{}_{main}+A_{slow}$ exp (-x/ $\tau_{slow}$) $^{\alpha}{}_{slow}$ protein when turns are excluded.

| Q | $y_0$ | $A_{main}$ | $\tau_{main}$ (ns) | $\alpha_{main}$ | $A_{slow}$ | $\tau_{slow}$ (ns) | $\alpha_{slow}$ |
|---|---|---|---|---|---|---|---|
| 1 | 0.00±0.00 | 0.59±0.10 | 0.18±0.23 | 0.35±0.05 | 0.38±0.21 | 13.91±4.53 | 0.88±0.30 |
| 1.5 | 0.065±0.13 | 0.54±0.27 | 0.13±0.23 | 0.28±0.06 | 0.36±0.33 | 14.31±7.15 | 0.90±0.43 |
| 2 | 0.00±0.08 | 0.6±0.22 | 0.05±0.04 | 0.28±0.08 | 0.31±0.21 | 11.23±3.16 | 0.87±0.35 |
| 2.5 | 0.00±0.00 | 0.63±0.10 | 0.05±0.01 | 0.26±0.04 | 0.19±0.05 | 8.99±0.38 | 0.99±0.13 |
| 3 | 0.00±0.00 | 0.69±0.19 | 0.01±0.00 | 0.22±0.06 | 0.12±0.06 | 6.98±0.22 | 0.98±0.25 |

Table S7- Fit parameters for calculated I (Q,t) of protein atoms of core with $y_0=y_0+A_{main}$ exp (-x/ $\tau_{main}$) $^{\alpha}{}_{main}+A_{slow}$ exp (-x/ $\tau_{slow}$) $^{\alpha}{}_{slow}$ protein.

| Q | $y_0$ | $A_{main}$ | $\tau_{main}$ (ns) | $\alpha_{main}$ | $A_{slow}$ | $\tau_{slow}$ (ns) | $\alpha_{slow}$ |
|---|---|---|---|---|---|---|---|
| 1 | 0.00±0.31 | 0.133±0.02 | 0.05±0.00 | 0.65±0.47 | 0.79±0.33 | 56.08±71.33 | 0.44±0.7 |
| 1.5 | 0.26±0.00 | 0.32±0.00 | 0.06±0.00 | 0.56±0.01 | 0.26±0.01 | 4.21±0.08 | 0.88±0.04 |
| 2 | 0.05±0.01 | 0.39±0.01 | 0.05±0.00 | 0.47±0.01 | 0.32±0.01 | 8.76±0.77 | 0.70(fix) |
| 2.5 | 0±0.05 | 0.48±0.03 | 0.06±0.01 | 0.37±0.02 | 0.22±0.07 | 11.83±3.50 | 1.00±0.22 |
| 3 | 0±0.01 | 0.36±0.02 | 0.025(fix) | 0.39±0.02 | 0.23±0.02 | 7.26±1.11 | 0.56±0.04 |

Table S8- Fit parameters for calculated I (Q,t) of protein atoms of the surface with $y_0=y_0+A_{main}$ exp (-x/ $\tau_{main}$) $^{\alpha}{}_{main}+A_{slow}$ exp (-x/ $\tau_{slow}$) $^{\alpha}{}_{slow}$ protein.

| Q | $y_0$ | $A_{main}$ | $\tau_{main}$ (ns) | $\alpha_{main}$ | $A_{slow}$ | $\tau_{slow}$ (ns) | $\alpha_{slow}$ |
|---|---|---|---|---|---|---|---|
| 1 | 0.00±0.12 | 0.25±0.01 | 0.07(fix) | 0.29±0.03 | 0.74±0.12 | 24.96±9.89 | 0.60±0.04 |
| 1.5 | 0.00±0.04 | 0.22±0.03 | 0.02±0.00 | 0.44±0.03 | 0.67±0.06 | 7.66±1.17 | 0.50±0.04 |
| 2 | 0.00±0.03 | 0.37±0.10 | 0.01±0.00 | 0.35±0.07 | 0.51±0.07 | 4.13±0.28 | 0.54±0.08 |
| 2.5 | 0.00±0.01 | 0.36±0.03 | 0.01±0.00 | 0.41±0.03 | 0.37±0.03 | 2.84±0.12 | 0.55±0.05 |
| 3 | 0.00±0.00 | 0.34±0.00 | 0.01±0.00 | 0.44(fix) | 0.28±0.00 | 1.81±0.06 | 0.55 (fix) |

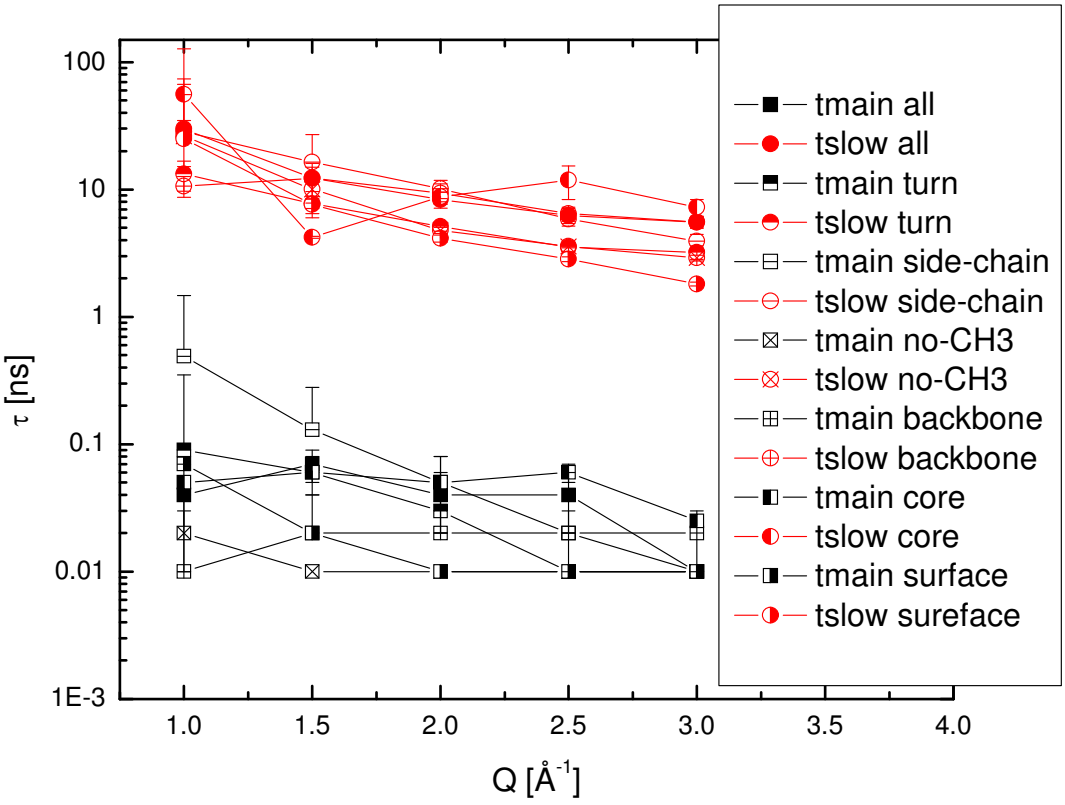


Fig S2. Relaxation times obtained from the fitting of the calculated I (Q,t) summarized from tables S1-S8.

Table S9- Fit parameters for $C_1(t)$ at different cut-off distance fitted by $y_0=y_0+A_1 \exp(-x/\tau_1)^{\alpha 1}+(1-(y_0+A_1)) \exp(-x/\tau_2)^{\alpha 2}$.

| D (Å) | $y_0$ | $A_1$ | $\tau_1$(ps) | $\alpha_1$ | $\tau_2$(ps) | $\alpha_2$ |
|---|---|---|---|---|---|---|
| 3 | 0.00±7.66 | 0.28±0.02 | 2.99±1.12 | 0.38±0.03 | 3592.45±45580.08 | 1.00±0.80 |
| 4 | 0.11±0.13 | 0.36±0.02 | 5.10±1.06 | 0.38±0.01 | 750.03±258.04 | 1.00±0.12 |
| 6 | 0.00±0.06 | 0.50±0.03 | 5.64±0.60 | 0.51±0.01 | 441.08±90.41 | 0.80±0.11 |
| 9 | 0.00±0.00 | 0.38±0.03 | 4.08±0.12 | 0.61±0.02 | 82.51±7.67 | 0.41±0.02 |
| All | 0.02±0.00 | 0.26±0.00 | 4.63±0.05 | 0.84±0.01 | 22.93±0.30 | 0.38±0.00 |

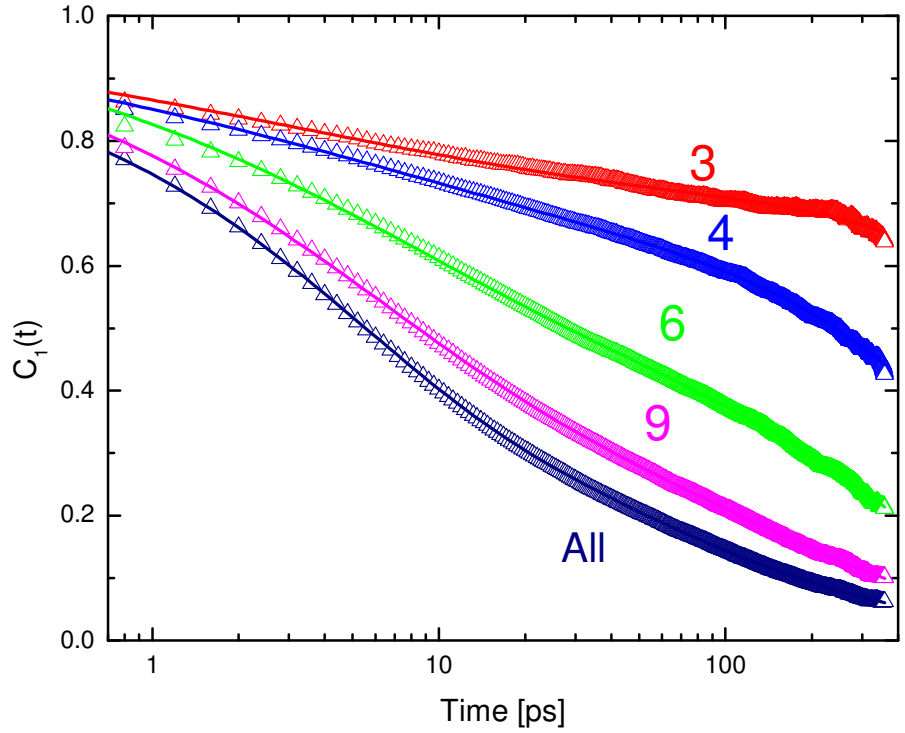


Fig S3. C1(t) of water molecule at different Cut-off distances (Å) indicated in the figure, symbols. The data have been fitted by $y_0=y_0+A_1 \exp(-x/\tau_1)^{\alpha 1}+(1-(y_0+A_1)) \exp(-x/\tau_2)^{\alpha 2}$, shown in lines. $R^2 > 0.97$.