Supplementary Material (ESI) for PCCP

Supplementary material to:

A Theoretical Spin Relaxation and Molecular Dynamics Simulation Study of the $Gd(H_2O)_9^{3+}$ Complex

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1 Supplementary material

In section **Generalized Solomon-Bloembergen-Morgan theory** Fig. 2 display the dipole-dipole correlation function, where the orientation of the vector from the metal ion and the water protons clearly display an approximative mono exponential decay.

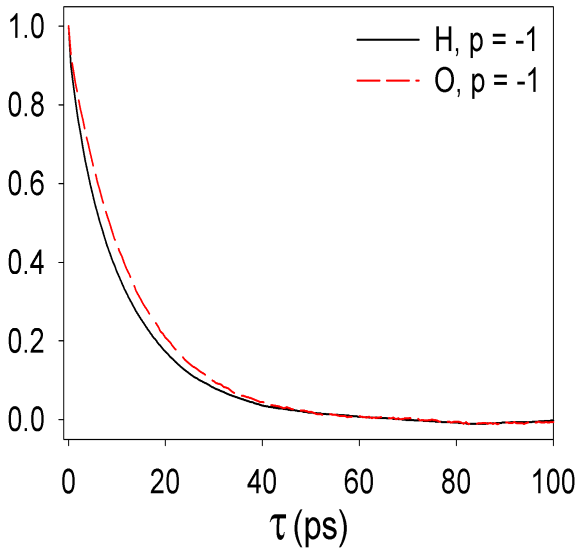

Figure 1: The normalized dipole-dipole correlation functions for the Gd-O and Gd-H vectors. The integral correlation times are $\tau_{Gd-H}/\tau_{Gd-O} = 0.87 \pm 0.03$

In Fig.(1) below, the dipole-dipole correlation functions of the dipole vectors defined from the metal ion to water or oxygen are displayed. Both dipole-dipole correlation functions are approximately mono exponential, but with slightly different correlation times: $\tau_{Gd-H}/\tau_{Gd-O} = 0.87 \pm 0.03$.

1.1 The origin of fast fluctuation in the electric field gradient calculation

The fast water flickering is observed for a single water molecule in the first hydration shell. However, this vibrational motion is averaged out for averages over long time or many water molecules. This is what happens for the dipole-dipole correlation function, but not for the electric field gradient calculation.

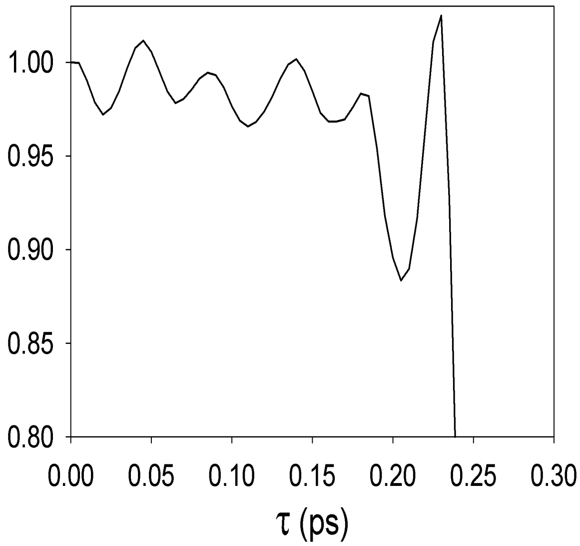

Figure 2: Example of a single molecules contribution to the normalized dipole-dipole correlation function for the hydrated metal complex

1.2 Dipole-dipole correlation function of different water models

For comparison the spin dipole-dipole correlation function was calculated from a Gd - SPC/E water simulation using the same Gd parametrization as in the TIP3P water simulation.

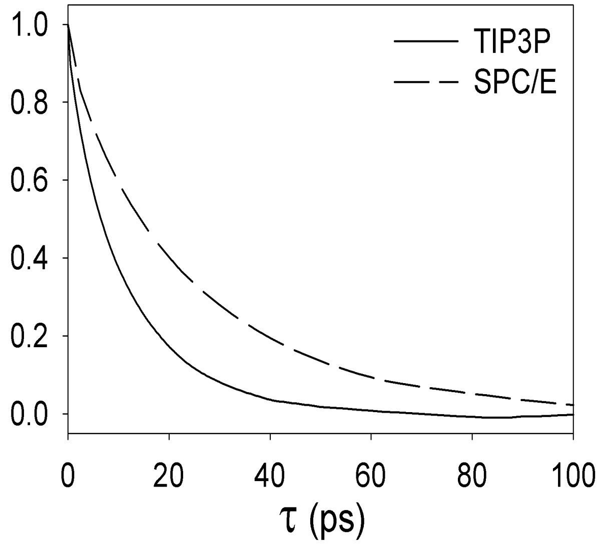

Figure 3: The Correlation function of SPC/E ($\tau_R = 23ps$) and TIP3P ($\tau_R = 10ps$) water for the hydrated metal complex