Quantum Chemistry Property Surface and machine learning in magnetic resonance relaxation modelling

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Interpretation of relaxation measurement can provide valuable information in chemistry, materials science and medicine for determining molecular structures, monitoring structural transformations, diagnostics and in probing dynamics at timescales relevant for biological systems [1]. Considering a long-lived nuclear singlet states (LLS), where a two-spin system is entirely correlated, have been shown to exceed magnetization (T_1) by a factor of 50, opening for new experiments [2]. However, in employing an analytical model of the relaxation a conclusive understanding is often challenging to reach due to a large set of unknown parameters.

An alternative to analytical relaxation model is the explicit simulation of spin-Hamiltonian trajectories. The spin Hamiltonian is built from NMR tensors that contain detailed information about the microstructure and dynamics of the samples. Accessible from first-principles quantum-chemical (QC) methods, the system-specific information is contained in the electronic structure of the substance. The relaxation times (T_1,T_{LLS}) are related to how the spin system couples to its environment. With QC sampling of the trajectory produced by atomistic molecular dynamics, relaxation observable may be computed directly, without adjustable parameters [3]. However, for accurate estimates of NMR relaxation, a vast number of QC calculations are needed, sampling many degrees of freedom (DOF) including both the spin moiety and the surrounding solvent, which makes the approach prohibitively costly.

We will present how this computational-bottleneck is overcome with development of Machine-learning QC-property surface. Central is the high-dimensional model representation [4], to manage the DOF. Application to LLS-relaxation due to the so-called spin-internal motion [2,3] will be illustrated.

References

1. J. Kowalewski and L. Mäler, *Nuclear Spin Relaxation in Liquids: Theory, Experiments, and Applications*, Taylor & Francis Group, New York, U.S.A., (2006).
2. G. Stevanato, J. T. Hill-Cousins, P. Håkansson, S. S. Roy, L. J. Brown, R. C. D. Brown, G. Pileio and M. H. Levitt, *Angew. Chem. (Int. ed.)*, **54**, 3740 (2015).
3. P. Håkansson, *Phys. Chem. Chem. Phys.*, **19**, 10237 (2017).
4. P. B. Nair and P. Håkansson, in *Proceedings of the 4th European Conference on Computational Mechanics (ECCSM IV)*, Paris, https://www.eccm-2010.org/abstract_pdf/abstract_1952.pdf, (2010).