Fast and accurate predictions of NMR parameters in aluminosilicate glasses via Machine Learning

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Nowadays, due to the intensive use of nuclear energy in our modern society, the treatment of radioactive wastes through vitrification becomes a thematic of central interest. In particular, the question of the efficiency, viability and durability resulting from such sequestration methods refers directly to our ability to understand and predict the structure and properties of these "nuclear glasses". Many simulation methods from MD, DFT to aiMD are used to investigate in this sense. Nevertheless, severe limitations exist such as the accuracy of the potentials used (MD) or the speed and feasibility of the calculations (aiMD). In the framework of our project, we aim to develop a MAChine Learning Approach for the pREdiction of NMR parameters for oxide glasses (MACLAREN). We propose to apply the state-of-the-art machine learning (ML) methods [1, 2] to: *(i)* use ML to provide a systematic approach for analyzing experimental NMR data (disentangle the atomic structure to NMR signals relationships), *(ii)* go beyond scalar predictions and predict different DFT observables such as total energy or atomic forces and *(iii)* accelerate the procedures already used in aiMD calculations (construct accurate ML force fields). The MACLAREN project subscribes in a global perspective aiming at the amelioration of modeling tools hitherto available for nuclear glasses studies. We plan to incorporate ML optimized schemes and the outputs from many spectroscopic approaches (NMR, RAMAN, structure factor measurements) into a reverse Monte-Carlo routine (fpNMR [3]) currently being developed. The aim is to have enough constraints to reach unprecedented accuracy in the establishment of the atomic structures of real samples (HRMC method, talk of T. Charpentier). We have already successfully designed ML procedures based on linear ridge regression [4] (LRR) combined with the smooth overlap of atomic positions [5] (SOAP) descriptors to predict the isotropic magnetic shielding (σ_{iso}) for different multicomponent relevant glass compositions. In our work, the ML predictions of σ_{iso} deviate, in best cases, from DFT-GIPAW calculations, by 0.7 ppm (as shown in Fig. 1) for ^{29}Si (1.0 % of the calculated σ_{iso} total span) and 1.5 ppm for ^{17}O (1.9 %) in SiO_2 glasses, 1.4 ppm for ^{23}Na (1.5 %) in Na_2O-SiO_2 and 1.5 ppm for ^{27}Al (2.1 %) in Al_2O_3-Na_2O-SiO_2 systems [6]. The ongoing developments are focused on the construction of ML force fields (part-*iii*) to validate the reliability of the simulated glass structures obtained via ML with those treated through the expensive aiMD simulations. With such ML schemes, many systems of high interest in the oxide glasses community, such as strontium aluminosilicate glasses [7] or borosilicate glasses [8], can be investigated much more efficiently.

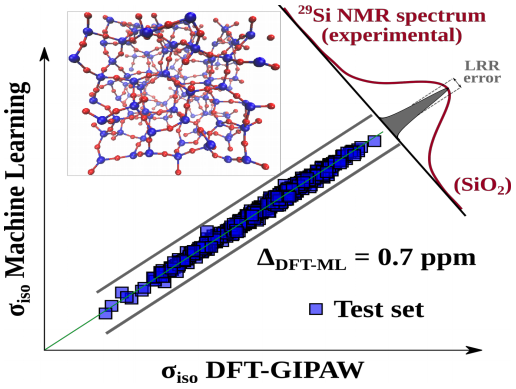

Fig. 1. Machine learning prediction of NMR isotropic magnetic shielding (σ_{iso}) for ^{29}Si nucleus in SiO_2 glass

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