

Towards exact first-principles predictions of spin-lattice relaxation in magnetic molecules and solid-state defects

Alessandro Lunghi

School of Physics, CRANN Institute and AMBER centre, Trinity College, Dublin 2, Ireland

email: lunghia@tcd.ie

Spin-lattice interaction is one of the main limitations to the spin lifetime in semiconductors and the control of such interaction is key for the development of technologies based on spin. Although spin-lattice relaxation has a central role in the physics of magnetism and magnetic resonance, its theoretical formulation is mostly based on a phenomenological approach and a quantitative understanding of its microscopic origin is often lacking.

In this contribution I will show the progresses in developing a computational approach able to tackle the challenge of predicting spin-lattice (T_1) relaxation from first principles. The formalism is based on a spin Hamiltonian description of magnetism and exploits the theory of open quantum systems in order to describe the dissipative effect of phonons on the spin degrees of freedom. This formalism is then mapped onto electronic structure theory, such as Density Functional Theory and Complete Active Space SCF, in order to determine all the parameters of the model in a full first-principles fashion[1].

Such computational strategy is shown to be able to quantitatively predict spin-phonon relaxation time in solid state compounds[1]. Results for both molecular crystals of Kramers ions and solid-state defects/impurities will be presented. The list of materials includes prototypical molecular qubits, such as $S=1/2$ V(IV) complexes[1], single-ion magnets, such as $S=3/2$ Co(II) and $J=15/2$ Dy(III) compounds[1-3], and color centers in diamond and hexagonal boron-nitride[4].

Ab initio spin dynamics is able to explain the dependence of spin-lattice relaxation time with respect to temperature for all the investigated compounds, making it possible to individuate the origin of slow relaxation. Moreover, we successfully disentangled all the spin and molecular interactions leading to spin-phonon coupling, making it possible to discuss future directions for this field and possible strategies for the enhancement of spin lifetime in solid-state spin systems

[1] A. Lunghi. *Science Advances*, 8, eabn7880 (2022)

[2] M. Briganti, et al. *The Journal of the American Chemical Society*, 143, 13633–13645 (2021)

[3] S. Mondal, A. Lunghi, *The Journal of the American Chemical Society*, 144, 22965-22975 (2022)

[4] S. Mondal, A. Lunghi, *npj Computational Materials* 9 (1), 120 (2023)