Eigenstates Engineering for Decoherence Mitigation in Molecular Spin Qudits

Giacomo Sansone[1,2], **Leonardo Ratini**[1,2], **Elena Garlatti**[1,2], **Francesco Petiziol**[3], **Paolo Santini**[1,2], **Stefano Carretta**[1,2]

1. *Dipartimento di Scienze Matematiche, Fisiche e Informatiche, Università di Parma, I-43124 Parma, Italy*
2. *Gruppo Collegato di Parma, INFN-Sezione Milano-Bicocca, I-43124 Parma, Italy*
3. *Technische Universität Berlin, Institut für Theoretische Physik, Hardenbergstr. 36, 10623 Berlin, Germany*

Abstract : A microscopic model of decoherence for molecular spin Qudits is developed. It shows that the decoherence time of a superposition of two eigenstates reflects differences between expectation values of local spins operators for those states.

Molecular Nanomagnets (MNMs) represent an attractive platform for quantum technologies, providing many low energy spin states that can be easily addressed through electromagnetic pulses to process quantum information [1]. This characteristic recently led to the suggestion of encoding Qudits within single molecules. At low temperatures, the primary error in these systems is pure dephasing, caused by interactions between the molecular spin and the surrounding nuclear spin bath. In recent years, significant efforts were devoted to bath engineering to reduce its impact on decoherence [2]. This approach led to important improvements in coherence times for molecular systems [3]. However, these achievements pertain mostly to molecular Qubits, where the long coherence time is associated with the superposition of a specific pair of the system eigenstates.

The present work proposes a shift in perspective regarding the understanding and mitigation of environment-induced decoherence. Rather than focusing on engineering the nuclear bath, we exploit the chemical tunability of the energy spectrum and eigenvectors of MNMs. We develop a microscopic model of decoherence at low temperatures for molecular spin systems undergoing pure dephasing. We report the outcome of analytical calculations highlighting the correlation between coherence time and the differences between expectation values of local spin operators evaluated on different eigenstates (these differences are identified with the Greek letter Δ in the figure below). These results are first tested upon a prototypical spin system, namely a single giant spin $S = 10$ (see Fig. 1 (a)), demonstrating that the lower the value of Δ, the better the coherence time. Then, a proposal for the realization of a new family of molecular Qudits is put forward: systems composed by six $S = 1/2$ spins characterized by competing (antiferromagnetic) interactions (Fig. 1 (b)). The theoretical approach exploited to simulate the coherence decay consists of a many-body technique known as Cluster Correlation Expansion (CCE) [4].

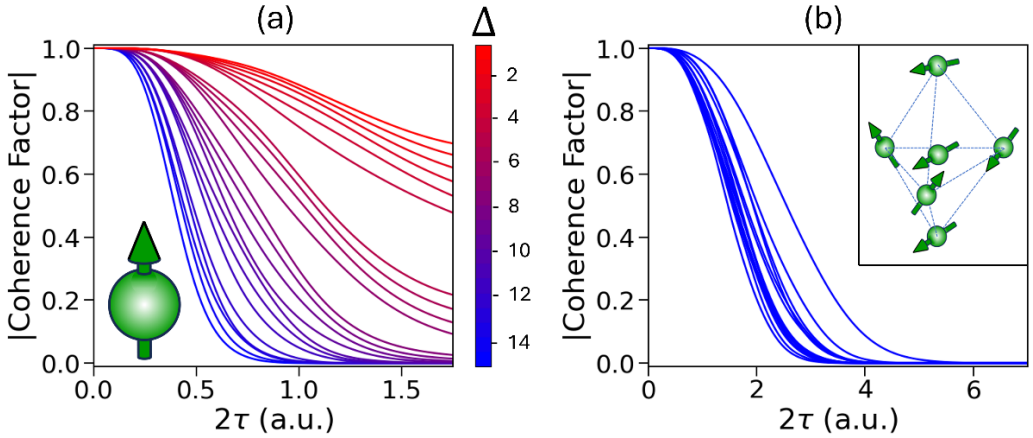

Fig. 1 (a) Decay of the coherence factor for different superpositions of the system eigenstates. The lower the value of Δ, the longer the coherence time (all the decay times are normalized with respect to the fastest decaying curve). (b) Proposal for a Qudit made of six spins ½ with competing interactions (inset). A set of eigenstates characterized by full connectivity through electromagnetic pulses allows the realization of many superpositions states with long coherence times. All the curves are plotted with respect to the decay time of an analogous system with ferromagnetic interactions.

References

[1] A. Chiesa, P. Santini, E. Garlatti, F. Luis, S. Carretta, "Molecular spins: a viable path toward quantum information processing?", Rep. Progr. Phys. **87**, 034501 (2024).

[2] M. J. Graham, C.-J. Yu, M. D. Krzyaniak, M. R. Wasielewski, D. E. Freedman, "Synthetic Approach To Determine the Effect of Nuclear Spin Distance on Electronic Spin Decoherence", J. Am. Chem. Soc. **139**, 3196 (2017).

[3] J. M. Zadrozny, J. Niklas, O. G. Poluektov, and D. E. Freedman, "Millisecond Coherence Time in a Tunable Molecular Electronic Spin Qubit", ACS Central Science **1**, 488 (2015).

[4] W. Yang, R.-B. Liu, "Quantum many-body theory of qubit decoherence in a finite-size spin bath", *Phys. Rev. B.*, **78**, 085315 (2008).