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CMQT Symposium

Guest Lecture Information

Stephen Hill

Stephen Hill currently holds the title of Distinguished Professor of Physics at Florida State University (FSU) with an affiliated appointment in the Department of Chemistry and Biochemistry, whilst also serving as Director of the Electron Magnetic Resonance user program at the United States National High Magnetic Field Laboratory (NHMFL). He previously held postdoctoral positions at Boston University and at the NHMFL, then took up faculty positions at Montana State University and the University of Florida before moving back to FSU in 2008. Hill has 30+ years of experience performing microwave and far-infrared magneto-optical and EPR spectroscopy of materials in high magnetic fields, using a wide array of compact radiation sources and measurement techniques. Through this work, he has gained an international reputation for spectroscopic investigations of low-dimensional conducting, superconducting and magnetic systems, including significant technique development. Hill's recent research has focused on fundamental studies of quantum phenomena in molecule-based magnets, as well as structure property relationships in a variety of inorganic coordination compounds. Hill was elected fellow of the American Physical Society (APS) in 2014, he won the Silver Medal for Instrumentation from the International EPR Society in the same year and then served in Chair's line of the APS Topical Group on Magnetism from 2016 to 2020. Most recently, he was elected to the Academy of Science, Engineering and Medicine of Florida (2024) and awarded the 2025 Royal Society of Chemistry Bruker Prize. Hill received both his Bachelors and Ph.D. degrees from the University of Oxford in the United Kingdom.

Molecular Hyperfine Clock Qubits

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Abstract: Recent work on molecular spin qubits has demonstrated significant enhancements in coherence through the engineering of so-called clock transitions, optimal operating points at avoided Zeeman level crossings where the quantum spin dynamics become desensitized to

magnetic noise [1]. The first examples focused on clock transitions generated by off-diagonal terms in the crystal-field Hamiltonian of integer spin-orbital (*J*) moment lanthanide ions (primarily Ho^{III}) in axial coordination environments – integer because the off-diagonal terms are even in angular momentum operators, therefore lifting the degeneracies within quasi-doublet, $m_J = \pm i$, projection states (i = integer). Unfortunately, the optimum coherence times (T_2) observed in such systems are severely limited by spin-lattice (T_1) relaxation due to the strong spin-orbit coupling (SOC) present in open-shell f-elements. This has motivated investigations of heavy elements with half-integer moments, where the clock transitions are generated instead via the off-diagonal part of the hyperfine interaction. In the case of lanthanides, a key trick involves reduction to the 2+ oxidation state which, for lutetium, results in a $4f^{14}(5d/6s)^1$ electronic configuration, i.e., a filled f-shell, with the extra electron occupying a mixed 5d/6s orbital, giving rise to a relativistically enhanced contact hyperfine interaction due to the s-orbital occupancy. Moreover, by increasing the overall s-orbital character, one can reduce SOC and enhance T_1 [2]. After an introduction, this talk will review recent efforts aimed at enhancing the s-orbital character in linear two-coordinate Lu^{II} complexes [3], as well as efforts targeted at generating hyperfine clock-transitions in other lanthanide 2+ complexes with $4f^n(5d/6s)^1$ electronic configurations [4], as well as several p-block elements including Bi and Tl.

[1] A. Gaita-Ariño, F. Luis, S. Hill, E. Coronado, Nature Chemistry **11**, 301 – 309 (2019).

[2] Kundu, Evans, Hill et al., Nature Chemistry **14**, 392 – 397 (2022).

[3] Ngo, Hrubý, Hill, Long et al, J. Am. Chem. Soc. (2025); https://doi.org/10.1021/jacs.5c01947

[4] Smith, Hrubý, Evans, Hill, Minasian, J. Am. Chem. Soc. **146**, 5781 – 5785 (2024).