Crystal-field modelling of hyperfine structure for low symmetry rare-earth doped insulators

Rare-earth doped insulators have over the past decade become key contenders in a variety of quantum information applications [1–9]. One of the primary areas in which rare-earth based systems have been highly successful is in long-term storage of quantum information; such a memory is a core component for both quantum computing and quantum communication technologies.

The key method that enables long coherence time of rare-earth based quantum memories is the ZEFOZ technique [10]. This technique uses a fine-tuned external magnetic field in order to quench, to first order, the dephasing induced by spin flips in the host material. Due to the large number of available magnetic field configurations, one requires an accurate model of the magnetic response and the hyperfine states of both the ground and the excited state in order to predict a suitable magnetic field orientation and strength. In practice, this is achieved by modelling the ground and a selected excited state using separate effective Hamiltonians, referred to as “spin Hamiltonians.” While the ground-state spin Hamiltonian can typically be determined using electron-paramagnetic resonance, for the excited-state spin Hamiltonian it is generally necessary to perform an exhaustive array of optical spectroscopy techniques. As a consequence, application of the ZEFOZ technique has to date been restricted to a total of three materials:
Pr^3+ :Y_2 SiO_5 [10], Pr^3+ :La(WO_4)_3 [11], and Eu^3+ :Y_2 SiO_5 [8].

In this talk I will outline a method for fitting a crystal-field Hamiltonian, which is an effective Hamiltonian of the complete 4f^N configuration, in order to obtain the detailed knowledge of the excited-state hyperfine structure. While modelling of the hyperfine structure using a crystal-field Hamiltonian is not uncommon, the host materials of interest in quantum information applications generally have very low point-group symmetry substitutional sites. This means than an accurate fit of the required parameters is very challenging for the materials of interest. As a consequence, the developed method augments a conventional crystal-field fit by additionally fitting to spin Hamiltonian parameter tensors. This means the crystal field parameters are optimized to reproduce the detailed magnetic and hyperfine response encoded in the spin Hamiltonian, which allows for a prediction of the excited state hyperfine structure.

[1] J. J. Longdell, M. J. Sellars, and N. B. Manson, Physical Review Letters 93, 130503 (2004). [2] M. P. Hedges, J. J. Longdell, Y. Li, and M. J. Sellars, Nature 465, 1052 (2010).
[3] C. Clausen, I. Usmani, F. Bussières, N. Sangouard, M. Afzelius, H. de Riedmatten, and N. Gisin, Nature 469, 508 (2011).
[4] G. Hétet, J. J. Longdell, A. L. Alexander, P. K. Lam, and M. J. Sellars, Physical Review Letters 100, 023601 (2008).
[5] S. A. Moiseev and S. Kröll, Physical Review Letters 87, 173601 (2001).
[6] M. Afzelius, C. Simon, H. de Riedmatten, and N. Gisin, Physical Review A 79, 052329 (2009).
[7] G. Wolfowicz, H. Maier-Flaig, R. Marino, A. Ferrier, H. Vezin, J. J. Morton, and P. Goldner, Physical Review Letters 114, 170503 (2015).
[8] M. Zhong, M. P. Hedges, R. L. Ahlefeldt, J. G. Bartholomew, S. E. Beavan, S. M. Wittig, J. J. Longdell, and M. J. Sellars, Nature 517, 177 (2015).
[9] C. Yin, M. Rancic, G. G. de Boo, N. Stavrias, J. C. McCallum, M. J. Sellars, and S. Rogge, Nature 497, 91 (2013).
[10] E. Fraval, M. J. Sellars, and J. J. Longdell, Physical Review Letters 92, 077601 (2004).
[11] M. Lovrić, P. Glasenapp, D. Suter, B. Tumino, A. Ferrier, P. Goldner, M. Sabooni, L. Rippe, and S. Kröll, Physical Review B 84, 104417 (2011).