# Generating and enhancing spin-light interfaces in molecular qubits

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Spins in molecules provide a platform for quantum information science which can deploy the versatility of synthetic chemistry. In particular, molecular ground-state electronic spins which can be interfaced with light are attractive as quantum bits (qubits). Such systems could combine key features of semiconductor spin qubits–e.g., single-spin detection and high-fidelity coherent control–with the versatility afforded by a molecular architecture, and therefore open up tailor-made qubits for applications such as nanoscale quantum sensing.

Here we outline our work to generate optical interfaces for ground-state molecular spins. Using Cr(IV)-based molecules, we show how core functionality of semiconductor spin qubits—optical initialization, optical read out, and coherent microwave control—can be realised in a molecular architecture [1]. We then outline two distinct ways in which the spin coherence of these optically addressable molecular qubits can be enhanced via control over the qubit's symmetry: (*i*) modifying the ligands co-ordinating the qubit, and (*ii*) modifying the qubit's host matrix [2]. These modifications induce magnetic-field insensitive spin transitions (clock transitions) which enhance coherence, and we model this behaviour from first principles across a series of molecules using cluster-correlation expansion methods. Finally, we show how host-matrix control can enhance other key qubit properties including optical spin initialisation and read out fidelities, and spin-lattice relaxation times.

Overall, these results demonstrate how core qubit properties of optical initialization, optical read out and coherent control can be realised in ground-state molecular spins, how the portability and tunability of molecular systems can be used to enhance qubit properties, and highlight opportunities for quantum sensing with optically active molecular spins.

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**References**

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