

Magnetic Control of Molecular Emission with Spin Qubit Pairs as the Basis of Quantum Sensing

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Photogenerated spin-correlated radical pairs (SCRPs) in electron donor–bridge–acceptor (D–B–A) molecules can act as molecular qubits and inherently spin qubit pairs. SCRPs can take singlet and triplet spin states, comprising the quantum superposition state. Coherence between two spin states and spin selective electron transfer reactions form the foundation of using SCRPs as qubits for sensing. We can exploit the unique sensitivity of the spin dynamics of SCRPs to external magnetic fields for quantum sensing applications including resolution-enhanced imaging, magnetometers, and magnetic switch [1]. Molecular quantum sensors, if realized, can provide new technological developments beyond what is possible with classical counterparts. We can use molecular emission, associated with spin-selective recombination of SCPRs, as a readout of spin state of SCRPs; optically “addressing” only one spin state of SCRPs.

Charge recombination to the electronic ground state typically occurs nonradiatively. We have demonstrated a rational design of donor-bridge-acceptor molecules that exhibit radiative charge recombination of radical pairs (charge-transfer emission) over long distances by taking advantage of intensity borrowing. Large excited-state electronic couplings and small energy gaps enable the observation of intramolecular long-range CT emission over unprecedented long distances. [2] Utilizing spin dynamics of radical pairs, we present a new strategy to control molecular emission by weak magnetic fields (< 1 Tesla), orders of magnitude smaller than the thermal energy at room temperature. We further demonstrated the tunability of the field response range by changing the energy levels of radical pairs; the field strength required to affect the emission, expressed in terms of exchange interactions,[3] experimentally validating, for spin-correlated radical pairs, the formalism developed by Anderson.[4] The implications of our study in quantum sensing applications will be discussed.

References

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