Scattering for Electronic Control of Molecular Spin Qubits

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Motivation

Molecular spin qubits (MSQs) formed from the electron spin degrees of freedom of a molecular system are a promising platform for QIP because they can be chemically tailored to achieve desired behavior and appear well suited for deploying at scale [Coronado, *Nat. Chem.,* 2019]. Although MSQs can be entangled via a Heisenberg-like exchange interaction [DiVincenzo, *Phys. Rev. A,* 1998], controlling the degree of entanglement experimentally by switching the interaction on and off presents a distinct challenge [Coronado, *Nat. Chem.,* 2019]. We have found that the introduction of a delocalized ancillary electron (DAE) scattering from the MSQs mediates the entanglement without the need for switchable interactions, while also introducing new degrees of freedom with which the experimenter can control the degree of entanglement between the MSQs.

Setup

Our scattering setup, sketched at right, consists of a single DAE in a 1D wire discretized into sites labeled by integer $j$and separated by lattice spacing a. The wire could be realized by a single wall carbon nanotube (SWCNT) or a silicon nanowire exhibiting ballistic transport. The left (right) lead is a noninteracting region of the wire consisting of identical sites $j \leq 0$ ($j < N$) where the DAE wavefunction is a plane wave. The scattering region consists of sites $j = 1, \ldots N$ where the DAE wavefunction is perturbed due interactions. We concentrate on the case of a single molecule, laterally coupled to the scattering region, which hosts two metal atoms (e.g. a dimer of single-molecule magnets). The spin degrees of freedom of the two metal atoms form two MSQs, which can interact with the DAE via an exchange interaction, represented by the on-site Hamiltonian

$$\langle j|H|j\rangle = J\vec{S}_e \cdot \left(\overrightarrow{S_1}\,\delta_{j1} + \vec{S}_2\delta_{j2}\right)/\hbar^2. \quad (1)$$

When $N = 2$, $\overrightarrow{S_1}\,\delta_{j1} + \vec{S}_2\delta_{j2} \approx \overrightarrow{S_1} + \vec{S}_2$ and the magnitude s_{12} of this combined spin operator is approximately conserved. As a result, the three particle spin states

$$|i\rangle \equiv |-1/2\rangle_e|s\rangle_1|s\rangle_2$$

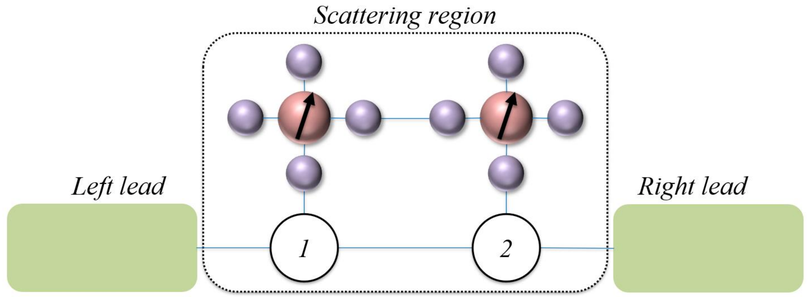
and

$$|+\rangle \equiv |+1/2\rangle(|s\rangle_1\,|s-1\rangle_2 + |s-1\rangle_1|s\rangle_2)/\sqrt{2}$$

are coupled exclusively to one another. Scattering from Eq. (1) has been previously studied in solid state systems [Viera, New J. Phys, 2006], but molecular systems are characterized by magnetic anisotropy and Heisenberg-like exchange, which allows inelastic scattering because the states $|i\rangle$ and $|+\rangle$ are split in energy by

$$\Delta E = (1-2s)D + s(J^x_{12} - J^z_{12}), \quad (2)$$

where s is the spin of each metal atom, D is its magnetic anisotropy, and J_{12} is the Heisenberg-like exchange interaction between them.

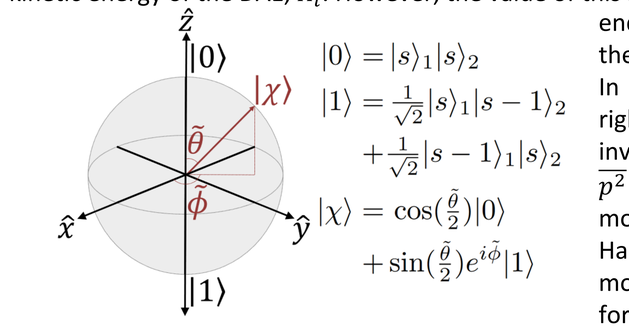


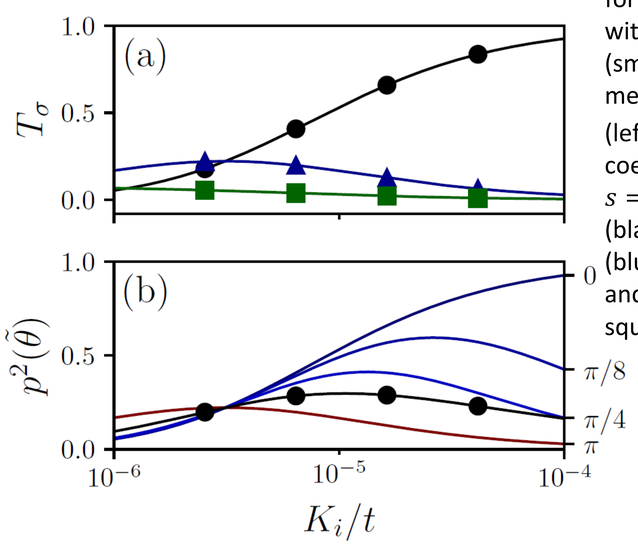
Control on the Bloch Sphere

Measurement of $\vec{S}_e$ projects the MSQs onto the Bloch sphere state $|\chi\rangle$ (defined in the figure below) probability given by

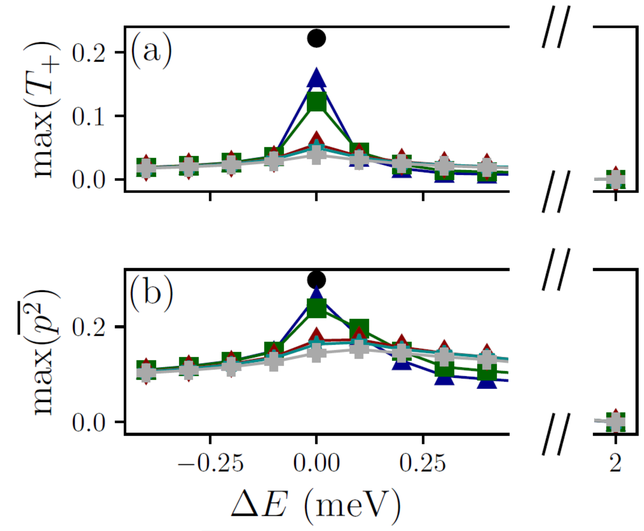
$$p^2 = T_i T_+ / [T_+ \cos^2(\tilde{\theta}/2) + T_i \sin^2(\tilde{\theta}/2)]$$

We are interested in maximizing p^2 and its average over $\tilde{\theta}, \overline{p^2}$. For a given molecule, this can be done straightforwardly by tuning the incident kinetic energy of the DAE, K_i. However, the value of this maximum depends intimately on the molecule itself. In the plots at right, we have investigated how $\overline{p^2}$ depends on the model Hamiltonian. This motivates a search for real molecules with desirable (small s, $\Delta E \ll 1$ meV) features.

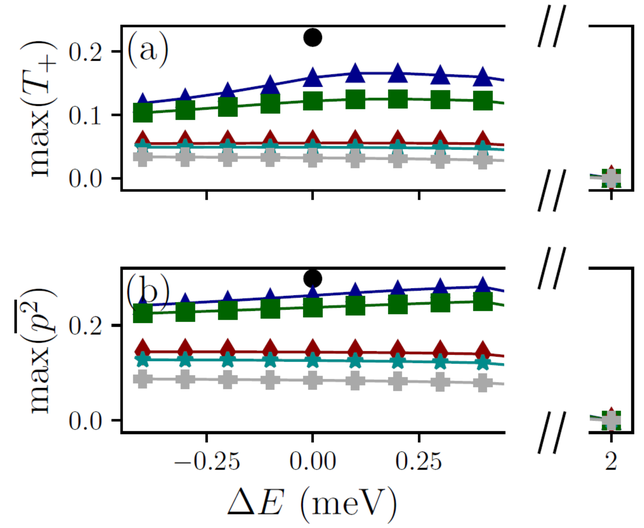




(left) transmission coefficients in the $s = 1/2$ case, with (black circles) T_i, (blue triangles) T_+ and (green squares) $T_- \times 10^5$.



Effects of ΔE and s on $\overline{p^2}$, where (black circles) $s = 1/2$, (blue triangles) $s = 1$, (green squares) $s = 3/2$, (red diamonds) $s = 4$, (cyan stars) $s = 9/2$, and (gray pluses) $s = 6$. (above) $J = -0.5$ meV as measured for $TbPc_2$ [Wernsdorfer, *Nature Mater.,* 2011], (below) $J = -5.0$ meV. J is determined by positing $2g\,\mu_B B_c = k_B T_K + J\mu_B S^z_{1,2}$ where g is the electron g-factor, B_c is the critical field, T_K is the Kondo temperature, and $S^z_{1,2}$ is the z-component of the MSQ spin, then measuring B_c and T_K [Wernsdorfer, *Nature,* 2012]. J likely depends very sensitively on the choice of molecule and how it is positioned on the wire, but this is not well studied.



Acknowledgements

This work was supported as part of the Center for Molecular Magnetic Quantum Materials, an Energy Frontier Research Center funded by the U.S. DOE/BES under Award No. DE-SC0019330.

https://doi.org/10.48550/arXiv.2210.15747