Scattering for Entangled State Switching in Molecular Dimers

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Motivation

Molecular dimers with two spin centers are a natural system for realizing entanglement, and a potential building block for quantum computation [1]. However, realizing experimental control of the entanglement presents a distinct challenge in molecular systems, where a linker must be employed as an on/off switch to control the exchange interaction between the spin centers. An alternative is to use a mediating particle to enable entanglement. A scattering process is a low control paradigm that defines the mediated entanglement process spatially, allowing the energy and momentum of the mediator to enter into the physics [2].

In existing electron mediated entanglement schemes, there is a need for direct contact with first principles descriptions of molecules [3]. In order to accomplish this, we employ a tight binding Green's function solution to the scattering problem. The tight binding framework naturally connects with the physical parameters of molecular qubits, enabling more realistic work. Furthermore, the idea of specializing electron mediated entanglement to molecular dimers has not been previously studied because the introduction of a mediating particle is typically motivated by the desire to have two well separated particles interact.

Method

We study a tight binding chain where the interactions are confined to a central region. The noninteracting, semi-infinite leads are accounted for via a self-energy term. Additional quantum numbers are introduced on each site to account for electron and impurity spin. This results in an effective Hamiltonian, with block matrices on each spatial site whose elements run over spin degrees of freedom.

$$\mathcal{H}' = \begin{pmatrix} \mathbf{H}_0 & \mathbf{t}_h & 0 & 0 \\ \mathbf{t}_h & \mathbf{H}_1 & \mathbf{t}' & 0 \\ 0 & \mathbf{t}' & \mathbf{H}_2 & \mathbf{t}_h \\ 0 & 0 & \mathbf{t}_h & \mathbf{H}_3 \end{pmatrix} + \begin{pmatrix} \mathbf{\Sigma}_0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \mathbf{\Sigma}_3 \end{pmatrix}$$

This effective Hamiltonian is used to obtain the Green's function of the system. Previous works have applied this method to finding spin polarized transport through systems of interest in nanoscale device physics, while treating the degrees of freedom on the impurity, junction, or interface as static [4]. Our work is the first to apply this method to the spin dependent scattering problem by including in the channels the spin states of the impurities rather than just the spin of the incident flux.

System

Our system (depicted in the next column) contains a mediating electron $\boldsymbol{S}_e$ and a molecule with two spin-s impurities $\boldsymbol{S}_1, \boldsymbol{S}_2$ located at sites j=1, j=2 respectively. Due to the $J\ \boldsymbol{S}\cdot\boldsymbol{S}$ interactions, the incident state $|k_i\rangle$ may be reflected into $|k_r\rangle$ or transmitted into $|k_t\rangle$. Recall that the elements of the Hamiltonian blocks $\mathbf{H}_j$ run over the spin states of the three particles, and the same is true of the $|k\rangle$ vectors.

The molecule consists of two sites connected by hopping t', which are connected to the leads via hopping t_h. The leads are taken to be semi-infinite tight binding chains with hopping t. Because we have discretized the system, we can use the parameters t_h, t', J_1, J_2 to parameterize the physics of the molecule of interest. The barrier on the right has height, width and position which are all experimentally tunable.

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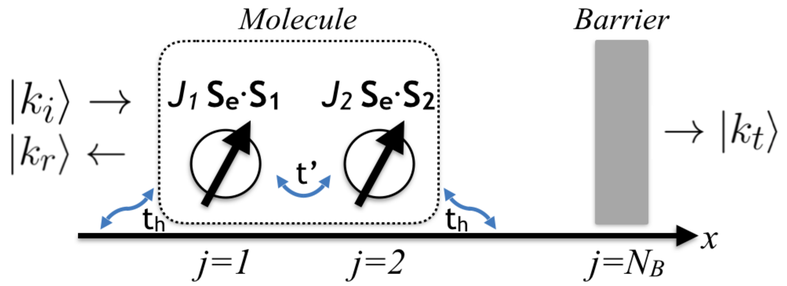


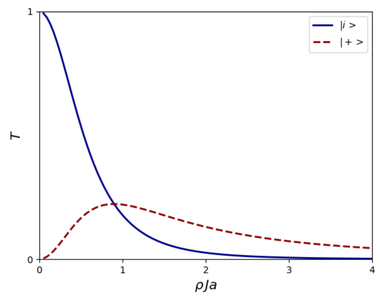
Entangled State Switching

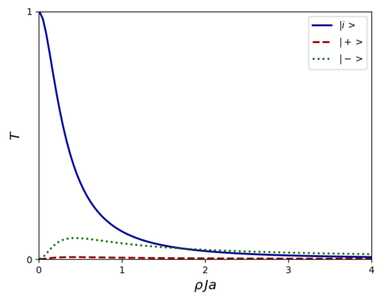
For the setup described above, we seek to couple an unentangled state $|i\rangle = |\downarrow\rangle_e|s\rangle_1|s\rangle_2$ to a maximally entangled state $|+\rangle = |\uparrow\rangle_e \frac{1}{\sqrt{2}}(|s\rangle_1|s-1\rangle_2 + |s-1\rangle_1|s\rangle_2)$. Then we can switch the entanglement of the system by transitioning between these states [3]. This is accomplished by the minimal Hamiltonian

$$\mathbf{H}_j = J_1\mathbf{S}_e\cdot\mathbf{S}_1\delta_{j1} + J_2\mathbf{S}_e\cdot\mathbf{S}_2\delta_{j2}$$

only if the electron couples identically to each impurity. Physically, this requirement is due to conservation of $S_1 + S_2$ which is necessary for entanglement switching.

In the left figure below, we show the probability of transmission into the entangled state for two spin-1/2 impurities mediated by an electron. The dimensionless quantity $\rho Ja \sim 1/k$.





Contact with Molecular Dimers

We used a toy model with J as a tunable parameter as a first realization of entangled state switching. A more complete treatment is obtained by using a density functional theory DFT parameterized Hamiltonian representing a Cobalt dimer:

$$\mathcal{H}_{Co2} = \mathbf{S}_O \begin{pmatrix} J_x & 0 & 0 \\ 0 & J_y & 0 \\ 0 & 0 & J_z \end{pmatrix} \mathbf{S}_T + D_O(S_O^z)^2 + D_T(S_T^z)^2$$

In the right figure above, we show the probability of transmission into the two possible entangled states for two spin-3/2 impurities mediated by an electron.

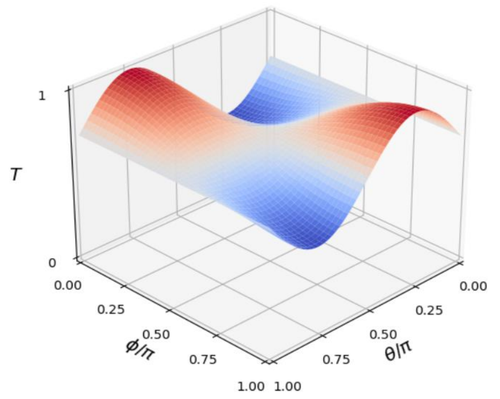
Entanglement Devices

Entangled State Detection

We prepare two spin-1/2 impurities in the generic entangled state parameterized by θ, ϕ:

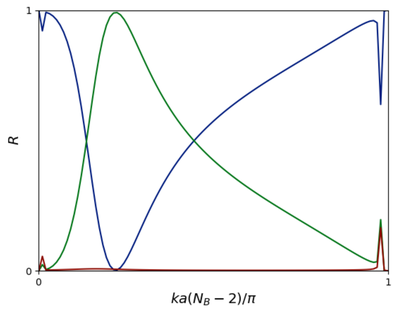
$$|\psi\rangle = \cos(\theta)|\uparrow\rangle_1|\downarrow\rangle_2 + e^{i\phi}\sin(\theta)|\downarrow\rangle_1|\uparrow\rangle_2$$

It turns out that the total transmissivity of the incident electron is very sensitive to the entanglement state of the molecular impurities. Spin singlets (e.g. $\theta = 3\pi/4, \phi = 0$) have maximum transmission while spin triplets (e.g. $\theta = \pi/4, \phi = 0$) have minimum transmission.



SWAP Gate

We prepare two spin-1/2 impurities in the state $|\uparrow\rangle_1|\downarrow\rangle_2$ and send in an incident electron $|\uparrow\rangle_e$ (blue line). The height and width of the barrier tend to infinity so that transmission is fully suppressed, so the reflection process becomes a unitary operation on the system [5]. By modulating the barrier position, we can SWAP into $|\uparrow\rangle_e|\downarrow\rangle_1|\uparrow\rangle_2$ (green line). At sufficiently low energies (pictured below), the spin-flip state $|\downarrow\rangle_e|\uparrow\rangle_1|\downarrow\rangle_2$ (red line) does not participate.



Conclusion

We have shown that entangling processes such as entangled state generation, entangled state detection, and entangling gates can be demonstrated using a tight binding solution that allows for direct contact with DFT. Our method allows us to go even further by replacing the spin parameters *J, D,* etc. with more fundamental spatial parameters such as molecular orbital energies and Coulomb charging energies, which would be interesting future work. Work towards scattering based control of the spin state of the cobalt dimer is also ongoing.

References

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