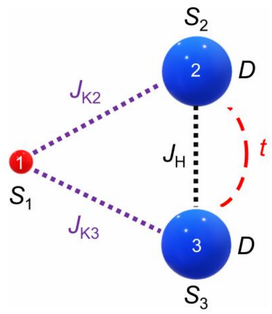
Spatial formulation of anisotropy exchange resonance

Christian Bunker,[1] Silas Hoffman,[1] Xiaoguang Zhang,[1] Garnet Kin-Lic Chan,[2] and Hai-Ping Cheng[1]
[1] Department of Physics, University of Florida, Gainesville, Florida 32611, USA
[2] Division of Chemistry and Chemical Engineering, California Institute of Technology, Pasadena, California 91125, USA

Motivation

Switzer et. al. [1] studied an effective spin only Hamiltonian for two spin-1 impurity sites with onsite spin anisotropy D_S interacting with an electron according to a Kondo like term J_K.



$$\mathcal{H}_A = D\left(\hat{S}_2^z\hat{S}_2^z + \hat{S}_3^z\hat{S}_3^z\right)$$

$$\mathcal{H}_{\mathrm{K}} = \frac{1}{2}\sum_{\mu,\mu',i=2,3} J_{\mathrm{Ki}}\hat{\mathbf{S}}_i\cdot\hat{\mathbf{d}}^\dagger_{\mu,i}\hat{\sigma}_{\mu,\mu'}\hat{\mathbf{d}}_{\mu',i}$$

They showed how certain values of D_S, J_K give rise to resonant transitions between fully entangled and non-entangled impurity states corresponding to different electron spins, so that the entanglement state of the system can be read out through the electron spin only.

$$|\downarrow\rangle|2,2\rangle = |\downarrow\rangle|1\rangle|1\rangle,$$
$$|\uparrow\rangle|2,1\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\rangle|0\rangle|1\rangle + |\uparrow\rangle|1\rangle|0\rangle\right),$$

We seek to derive their spin Hamiltonian from a fuller formulation with spatial degrees of freedom.

Methods

We consider a model Hamiltonian that encapsulates the physics of a fixed l molecular subshell by input parameters α for spin orbit coupling, D, E for spatial anisotropy, and U for Coulomb repulsion.

$$\mathcal{H} = 2\alpha\mathbf{L}\cdot\mathbf{S} + U\sum_{m=-l}^{l} n_{m\uparrow}n_{m\downarrow} + \mathbf{L}\cdot\begin{pmatrix} D/3+E & 0 & 0 \\ 0 & D/3-E & 0 \\ 0 & 0 & -2D/3 \end{pmatrix}\cdot\mathbf{L}$$

In this formulation D represents a z axis anisotropy and E an x-y plane anisotropy. We seek to recover the effective spin interaction terms in the appropriate limits of these inputs. Physically we would expect that $\alpha \ll E \ll D \ll U$.

Analytical Treatment

To derive an onsite spin-1 anisotropy term analytically, we look for electrons on the impurity combining such that <S> = 1, with an energy difference between the <S_z> = ± 1 and <S_z> = 0 states. Although many <S> = 1 combinations are possible, the time reversal symmetry of the system means that we are interested in time reversal partner states, i.e. states where $m \to -m, m_s \to -m_s$. Then Kramer's theorem guarantees degeneracy of these partner states.

For a 3-level system ($l = 1$) with 4 electrons, the Hamiltonian may be diagonalized by hand. Downfolding (which assumes $\alpha \ll D, E \ll U$) yields an effective Hamiltonian for the subspace of interest

$$\mathcal{H}_{eff} = \begin{pmatrix} -2\alpha - \frac{8E^2}{U} & \frac{8E^2}{U} & 0 & 0 \\ \frac{8E^2}{U} & 2\alpha - \frac{8E^2}{U} - \frac{2\alpha^2}{\alpha-D} & 0 & 0 \\ 0 & 0 & \frac{\alpha^2}{\alpha+D} & 0 \\ 0 & 0 & 0 & \frac{\alpha^2}{\alpha+D} \end{pmatrix} \begin{matrix} |\uparrow\rangle|\uparrow\downarrow\rangle|\downarrow\rangle \\ |\downarrow\rangle|\uparrow\downarrow\rangle|\uparrow\rangle \\ |\uparrow\rangle|\uparrow\downarrow\rangle|\uparrow\rangle \\ |\downarrow\rangle|\uparrow\downarrow\rangle|\downarrow\rangle \end{matrix}$$

Then we calculate the eigenstates and energies directly. The limits $\alpha/D \ll 1, E/U \ll 1$ are immediately applied. To recover spin-1 states, the eigenstates are considered orders of the small quantity $\alpha U/4E^2$ after a shift of $U - 2D$.

$$|S\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\rangle|\uparrow\downarrow\rangle|\downarrow\rangle - |\downarrow\rangle|\uparrow\downarrow\rangle|\uparrow\rangle\right) \qquad E_S = -\frac{16E^2}{U}$$
$$|T_0\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\rangle|\uparrow\downarrow\rangle|\downarrow\rangle + |\downarrow\rangle|\uparrow\downarrow\rangle|\uparrow\rangle\right) \qquad E_{T0} = \frac{\alpha^2}{4E^2/U}$$
$$|T_+\rangle = |\uparrow\rangle|\uparrow\downarrow\rangle|\uparrow\rangle \qquad E_{T+} = \frac{\alpha^2}{D}$$
$$|T_-\rangle = |\downarrow\rangle|\uparrow\downarrow\rangle|\downarrow\rangle \qquad E_{T-} = \frac{\alpha^2}{D}$$

Numerical Treatment

Using the PySCF quantum chemistry package [2], the full second quantized Hamiltonian may be numerically diagonalized for arbitrary values of α, E, D, U, for any number of basis sites or electrons. This allows us to consider a d orbital (5-level) impurity exactly. We confirm that the analytical results presented above hold in the limits $\frac{\alpha}{D}, \frac{E}{U} \ll \frac{\alpha U}{4E^2} \ll 1$ after a shift of $3U - 18D$. Thus we identify an effective onsite spin anisotropy

$$\mathcal{H}_A = D_S S_z^2, \quad D_S \equiv -\frac{\alpha^2}{4E^2/U}$$

$$J_L(t) = -it_{\mathrm{hyb}}\sum_\sigma \langle a^\dagger_{L1\sigma}a_{d\sigma} - a^\dagger_{d\sigma}a_{L1\sigma}\rangle$$

$$J_R(t) = -it_{\mathrm{hyb}}\sum_\sigma \langle a^\dagger_{d\sigma}a_{R1\sigma} - a^\dagger_{R1\sigma}a_{d\sigma}\rangle$$

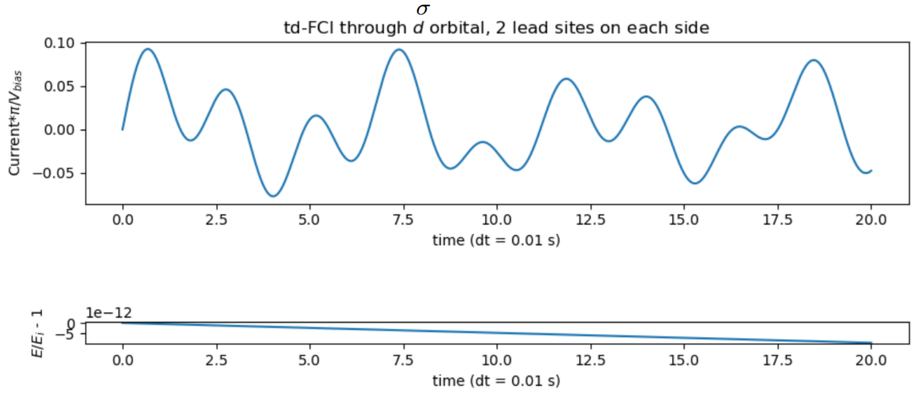


Figure 1: Current through half filled d orbital impurity model. Bias is turned on at time zero to introduce dynamics. Physical inputs $t = 1.0$, $t_{hyb} = 0.4$, $V_{bias} = -0.005$, $U = 1.0$, $\alpha = 0.01$, $E = 0.1$, $D = 0.5$.

The numerical treatment also opens the possibility of extending the model to a dynamic treatment. To do so, we implement the Single Impurity Anderson Model (SIAM) in which the impurity is connected to a chain of tight binding sites (leads). In other words [3],

$$\hat{H} = \hat{H}_{\mathrm{dot}} + \hat{H}_{\mathrm{leads}} + \hat{H}_{\mathrm{dot\text{-}leads}} + \hat{H}_{\mathrm{bias}}$$

where H_{dot} contains the physics of the impurity, (so here represents the model molecular Hamiltonian already introduced), H_{leads} allows hopping t between lead sites, and $H_{dot-leads}$ allows hopping t_{hyb} onto the impurity. A small bias between left lead sites and right lead sites, turned on at time zero, imposes nonequilibrium. Fig. 1 shows current through the impurity, prepared in the ground state, over time as a proof of concept.

Next Steps

The dynamic treatment afforded by SIAM formalism opens many new doors. In the spirit of Switzer's paper [1], we are interested in reading out the initial spin state of an impurity. This can be done within the SIAM framework by turning on, then off, a magnetic field like term H_{prep} that prepares the spin state of the impurity before time zero. Then we can investigate how dynamical properties such as current frequency, conductance, etc. are affected.

At the moment, the simulations are restricted to two site leads by the computational limits of td-FCI. To achieve a more realistic representation of an infinite lead, we will need to reproduce our results in the td-DMRG framework which can handle many more sites.

Finally, an extension of the analytical treatment to two molecular impurities in order to recover Kondo exchange is needed to rederive the results of [1].

References

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