Spin inelastic currents in molecular ring junctions

Dhurba Rai and Michael Galperin

Department of Chemistry and Biochemistry, University of California San Diego, La Jolla, CA

Abstract

We present a study of spin inelastic currents in molecular ring junctions by generalizing considerations of the spin-flip inelastic electron tunnelling spectroscopy (IETS) to the case of multisite molecular system and formulate a conserving approximation, taking into account renormalization of elastic channel. Within a simple model of a benzene molecule coupled to paramagnetic contacts at meta, ortho, and para positions, we demonstrate the role of external magnetic field and local spin impurity placed at the center of the ring on the control of spin-flip IETS signal, and present spin polarization of circular and total currents.

Model and Method

Model Hamiltonian

$$\hat{H} = \hat{H}_M + \sum_{K=L,R} \hat{H}_K + \hat{V}_{KM} + \hat{V}_{SM} \equiv \hat{H}_0 + \hat{V}_{SM} \qquad (1)$$

$$\hat{V}_{SM} = \sum_{m_1,m_2 \in M, \sigma_1,\sigma_2} J_{m_1 m_2} (\hat{\vec{S}} \cdot \vec{\sigma}_{\sigma_1\sigma_2}) \hat{d}^\dagger_{m_1\sigma_1} \hat{d}_{m_2\sigma_2} \qquad (2)$$

$$\hat{H}_K = \sum_{k \in K,\sigma} \alpha_K \hat{c}^\dagger_{k\sigma} \hat{c}_{k\sigma} + \sum_{\langle k_1,k_2 \rangle \in K,\sigma} \left(\beta_K \hat{c}^\dagger_{k_1\sigma} \hat{c}_{k_2\sigma} + \text{H.c.} \right) \qquad (3)$$

$$\hat{V}_{KM} = \sum_{\sigma} \left(\beta_{KM} \hat{c}^\dagger_{k_K\sigma} \hat{d}_{mK\sigma} + \text{H.c.} \right) \qquad (4)$$

$$\hat{V}_{SM} = \sum_{m_1,m_2 \in M, \sigma_1,\sigma_2} J_{m_1 m_2} (\hat{\vec{S}} \cdot \vec{\sigma}_{\sigma_1\sigma_2}) \hat{d}^\dagger_{m_1\sigma_1} \hat{d}_{m_2\sigma_2} \qquad (5)$$

where the notation have usual meanings. The onsite energy and intersite coupling strength are function of total (external plus induced) magnetic field/flux,

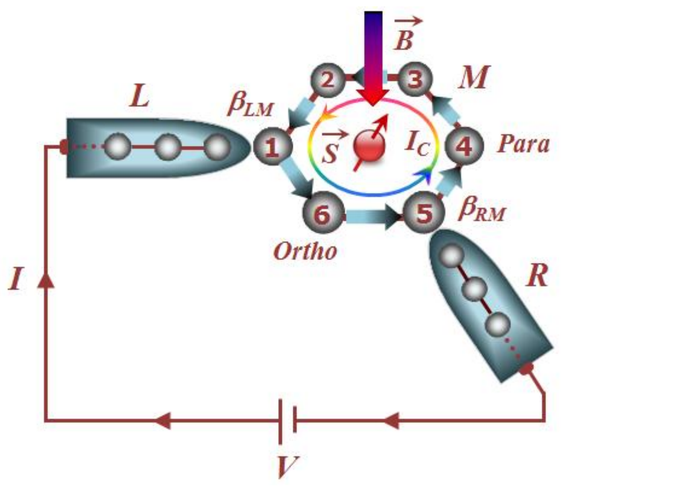
$$\alpha_{M\sigma} \equiv \alpha_M - 2\mu_B B_{\text{tot}} \sigma \qquad \beta_M \to \beta_M e^{i\theta} \qquad \theta = 2\pi \frac{\phi_{B_{\text{tot}}}}{6\phi_0} \qquad (6)$$

The magnetic field removes the degeneracy of the local spin eigenstates,

$$E_{SM_S} = -2\mu_B B_{\text{tot}} M_S \qquad (7)$$

The spin exchange interaction is modeled as

$$J_{m_1 m_2} = \delta_{m_1,m_2} J \quad \text{or} \quad J_{m_1 m_2} = \delta_{\langle m_1,m_2 \rangle} J \qquad (8)$$



Parameters Used

$\alpha_M = -2\,\text{eV},\ \beta_M = 2.5\,\text{eV}$

$\alpha_K = 0,\ \beta_K = 6\,\text{eV}\ (K = L, R)$

$\beta_{LM} = \beta_{RM} = 0.3\,\text{eV}$

$\Gamma_K = 2|\beta_{KM}|^2 / |\beta_K| = 30\,\text{meV}$

$S = 1,\ J = 5\,\text{meV},\ T = 0.5\,\text{K},\ E_F = 0$

and

$\mu_{L,R} = E_F \pm V/2.$

Fig. 1.Tight binding model for current conduction through a molecular ring *M* with spin impurity *S* at its center coupled to metal leads (*L*,*R*). External uniform field *B* is applied perpendicular to the ring plane.

Self Consistent Current Calculation

We employ non equilibrium Green's function NEGF) technique for current calculation. Spin-spin exchange interaction is treated as perturbation to the system described by free electron Green's function that incorporates the self-energies due to coupling of molecule to the leads. The second order perturbative expansion of Green's function in the spin-spin exchange interaction leads to the Dyson equation,

$$G_{mm',\sigma}(\tau,\tau') = G^{(0)}_{mm',\sigma}(\tau,\tau') + \sum_{m_1 m_2 \in M} \int_c d\tau_1 \int_c d\tau_2 G^{(0)}_{mm_1,\sigma}(\tau,\tau_1) \Sigma^{(S)}_{m_1 m_2,\sigma}(\tau_1,\tau_2)\, G_{m_2 m',\sigma}(\tau_2,\tau') \qquad (9)$$

with self-energy

$$\Sigma^{(S)}_{m_1 m_2,\sigma}(\tau_1,\tau_2) = \delta(\tau_1,\tau_2) \Sigma^{(S)\delta}_{m_1 m_2,\sigma} + \Sigma^{(S)el}_{m_1 m_2,\sigma}(\tau_1,\tau_2) + \Sigma^{(S)inel}_{m_1 m_2,\sigma}(\tau_1,\tau_2) \qquad (10)$$

The superscript (0) in Eq. (9) denotes the non-interacting Green's function. Each term in Eq. (10) depends on spin exchange strength (*J*) and on the probability of occupation of the spin level from which the magnetic field dependence enters. Green's function enters from the elastic (*el*) and inelastic (*inel*) terms. Equations (9) and (10) are solved self consistently. Self consistency results from the interdependence of Green's function, self energy and magnetic field induced by a circular current in the ring. The converged Green function is then used for current calculation through the following expressions,

(a) Intersite current, i.e., bond current

$$I^\sigma_{m_1 \to m_2}(t) \approx \frac{2e}{\hbar} \text{Re}\left[\beta_{m_1 m_2,\sigma} G^<_{m_2 m_1,\sigma}(t,t) \right]$$

(b) Circular current (defined as the sole source of magnetic flux through the ring)

$$I^\sigma_c(t) = \sum_{\langle m_1,m_2 \rangle \in M} I^\sigma_{m_1 \to m_2}(t) \frac{l_{\langle m_1,m_2 \rangle}}{\ell} \qquad \ell = \sum_{\langle m_1,m_2 \rangle \in M} l_{\langle m_1,m_2 \rangle} \qquad \beta_{m_1 m_2,\sigma} \equiv \beta_M + \Sigma^{(S)\delta}_{m_1 m_2,\sigma}$$

(Sum of bond lengths) (Effective intersite coupling strength)

(c) Terminal current

$$I^\sigma_K(t) = \frac{2e}{\hbar} \sum_{m_1,m_2 \in M} \text{Re} \int_{-\infty}^{t} dt_1 \left[\Sigma^{(K)<}_{m_1 m_2,\sigma}(t,t_1) G^>_{m_2 m_1,\sigma}(t_1,t) - \Sigma^{(K)>}_{m_1 m_2,\sigma}(t,t_1) G^<_{m_2 m_1,\sigma}(t_1,t) \right]$$

Spin polarization: $P = (I_\uparrow - I_\downarrow)/(I_\uparrow + I_\downarrow) = \eta_\uparrow - \eta_\downarrow$

Results

Steady state calculations are performed for 0.3 eV metal-molecule coupling strength and on the energy grid -1.5 eV to 1.5 ev in steps of 0.01 meV. Both spin exchange models (Eq. 8) give qualitatively same result. The main results are shown below

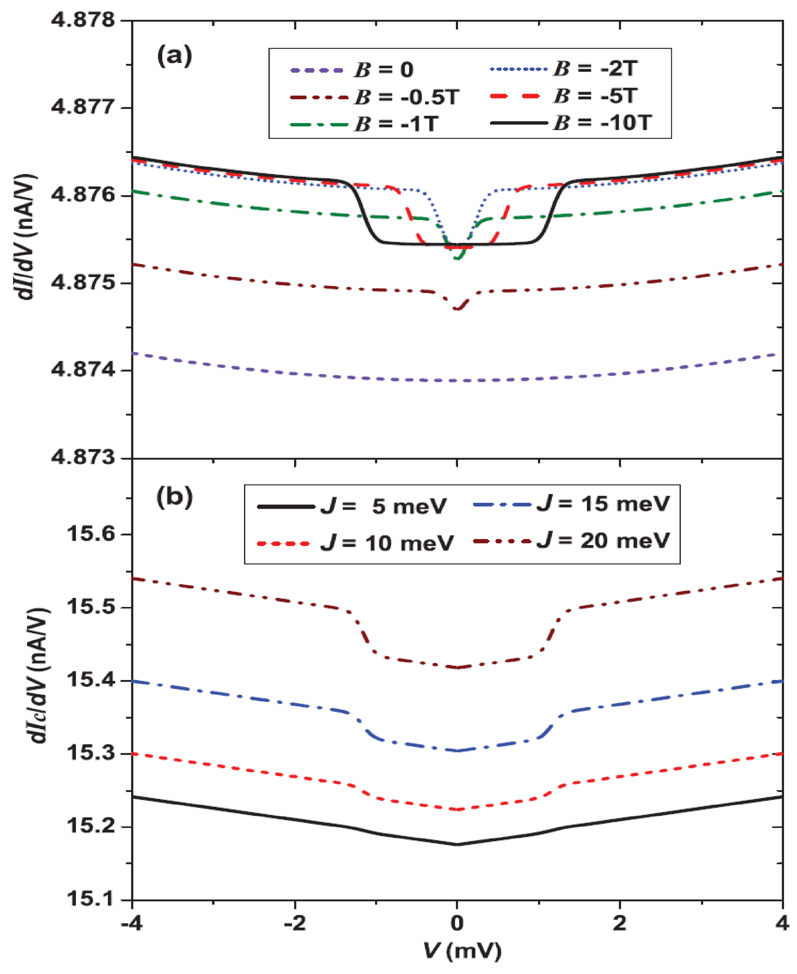


Fig. 2. dI/dV in meta connected ring for (a) terminal current at *J*=5 meV and (b) circular current at *B*=-10T.

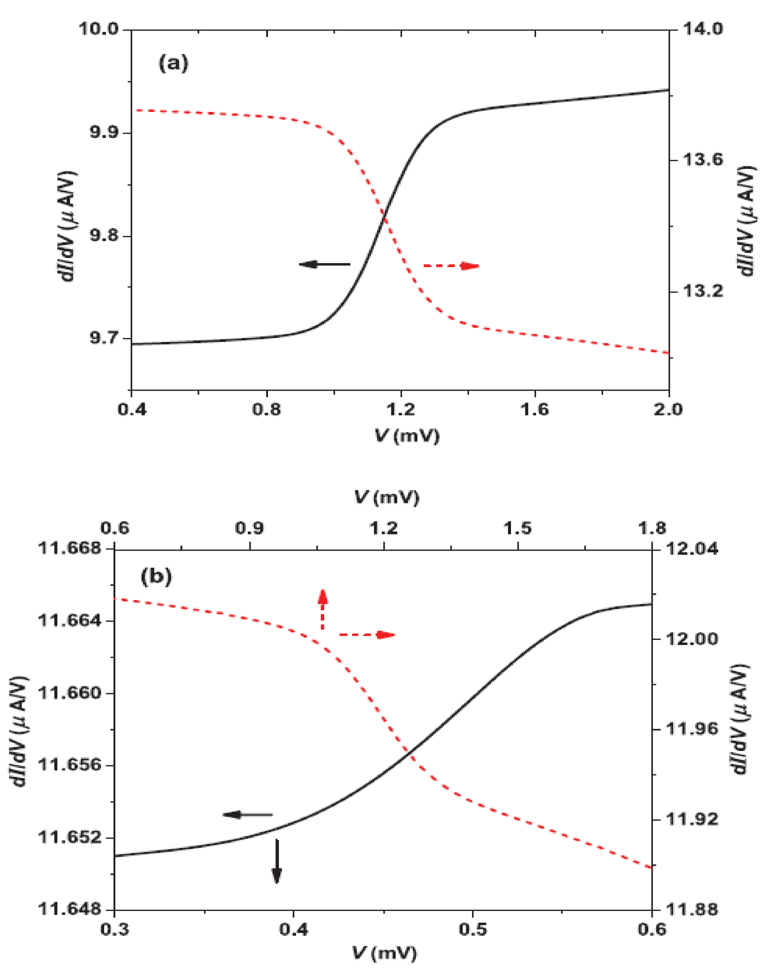


Fig. 3. dI/dV in meta connected ring at (a) *B*=-10T for Vg=0.486 V (solid line, black) and 0.490V (dashed line, red), (b) Vg=0.488 V for *B*=-5T (solid black line; left and bottom axes) and -10T (dashed red line; right and top axes). *J*=5meV.

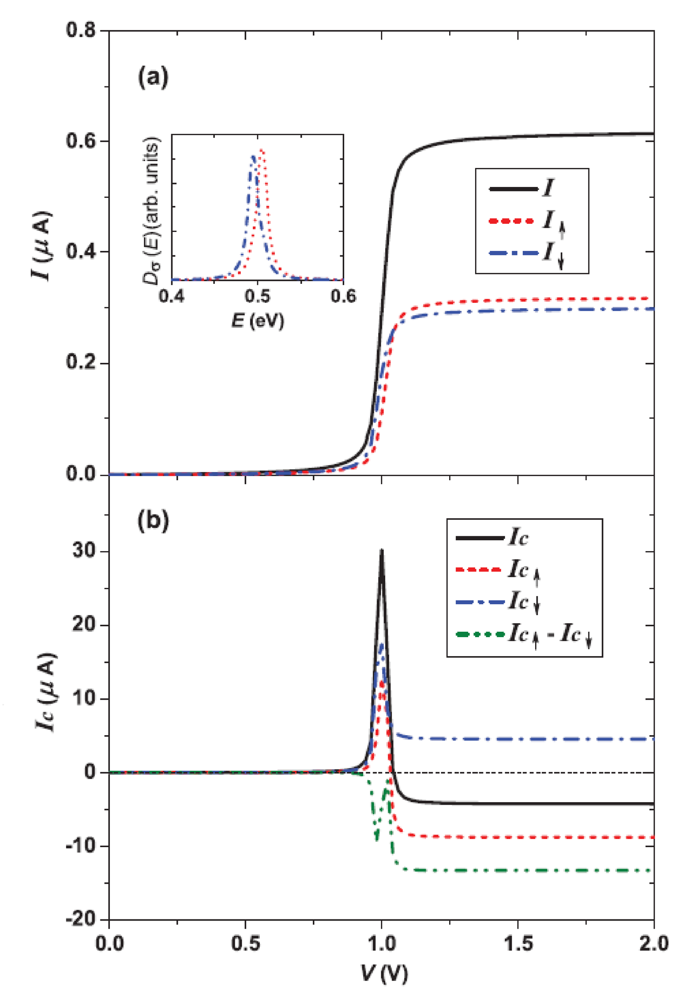


Fig. 4. (I-V) characteristics in meta connected ring for (a) terminal current and (b) circular current at *B*=-5T. The inset in (a) shows spin resolved density of states at 1V bias.

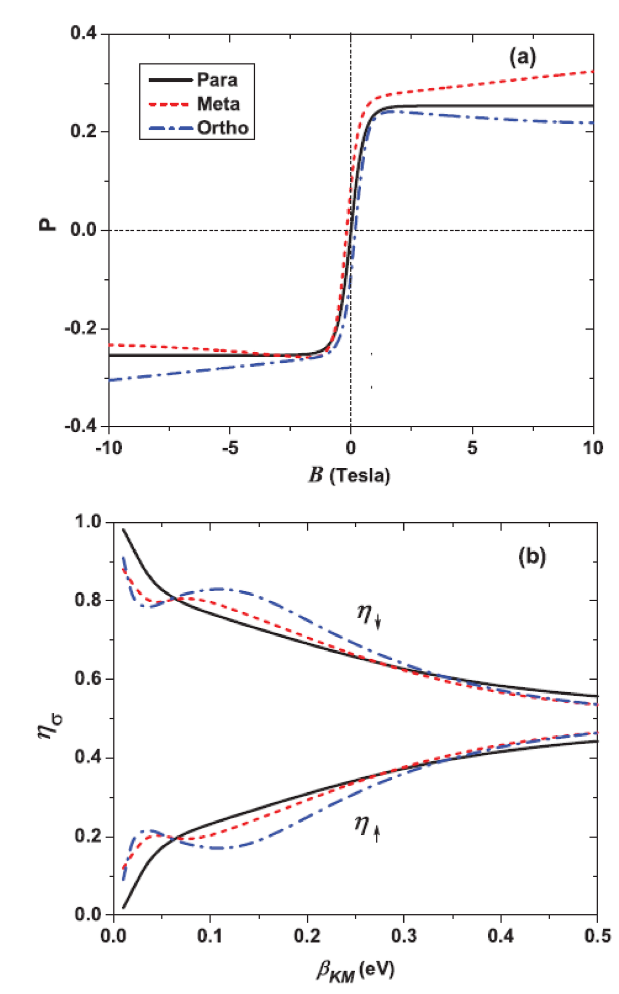


Fig. 5. (a) Spin polarization and (b) Spin filter efficiency for *B*=-5T.

Conclusion

1. **Like in vibrational IETS, spin-flip IETS yields the possibility of control of the IETS signal.**
2. **In addition to gate voltage, magnetic field can be used as a control of the spin-flip IETS spectrum in any junction with spin-spin exchange interaction.**
3. **Spin-spin exchange interaction in ring structures results in spin circular currents and such molecular rings can be used as sources of spin-polarized terminal currents.**
4. **For a given magnetic field and lead-molecule configuration, spin filter efficiency can be controlled by lead-molecule coupling strength.**

Acknowledgments

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