Spin inelastic currents in molecular ring junctions

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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 multilete 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

\[ H = H_f + \sum_{k} H_{\psi k} + \sum_{J} H_{V J} + \sum_{J} H_{c J} \] (1)

\[ V_{\psi k} = -\sum_{J} V_{\psi k} (S^J \psi_k^+ \psi_{k'}^J) \] (2)

\[ H_{V J} = \sum_{J} \left( J_{V J} \psi_k^J \right) \] (3)

\[ V_{c J} = \sum_{J} \left( V_{c J} \psi_k^J \right) \] (4)

\[ \alpha_{eff} = -2\mu B \sigma \] (6)

where the notation have usual meanings. The onsite energy and inter site coupling strength are function of total (external plus induced) magnetic field, \( \alpha_{eff} = -2\mu B \sigma \).

Self Consistent Current Calculation

We employ non equilibrium Green's function (NEGF) technique for current calculation. 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-exchange interaction leads to the Dyson equation.

\[ G_{\sigma}(\tau,\tau') = G_{\sigma}^{el}(\tau,\tau') + \sum_{\sigma'} \sum_{\alpha} \int d\tau_{\sigma} G_{\sigma'}(\tau,\tau_{\sigma}) G_{\sigma'}(\tau_{\sigma},\tau') \] (9)

with self-energy

\[ \sum_{\sigma'} G_{\sigma}(\tau,\tau') = \delta_{\sigma}\tau_{\tau} \sum_{\alpha} \delta_{\alpha}\tau_{\alpha} + \sum_{\alpha} G_{\sigma}(\tau_{\alpha},\tau_{\beta}) + \sum_{\alpha} G_{\sigma}(\tau_{\alpha},\tau_{\alpha}) \] (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 (\( J \)) and inelastic (\( \alpha \)) terms. Equations (9) and (10) are solved self-consistently. Self consistency results from the independence 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

\[ I_{\sigma_{\alpha \beta}}^{\alpha_0}(t) = \frac{2e}{h} \text{Re} \left[ \beta_{\sigma_{\alpha \beta}} G_{\sigma_{\alpha \beta}}(t, t) \right] \]

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

\[ I_{\alpha_0}^{e}(t) = \sum_{\sigma_{\alpha_\beta}} \text{Re} \left[ \frac{e}{h} \sum_{t, t'} \left( \frac{G_{\sigma_{\alpha_\beta}}(t, t')}{\epsilon - G_{\sigma_{\alpha_\beta}}^{el}(t, t')} \right) \right] \]

(c) Terminal current

\[ I_{\alpha_0}^{e}(t) = \frac{2e}{h} \sum_{\sigma_{\alpha_\beta}} \text{Re} \left[ \frac{e}{h} \sum_{t, t'} \left( \frac{G_{\sigma_{\alpha_\beta}}(t, t')}{\epsilon - G_{\sigma_{\alpha_\beta}}^{el}(t, t')} \right) \right] \]

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 eV. 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 0.6 meV and (b) circular current at Bi-10T.

Fig. 3. dI/dV in meta-connected ring at (a) Bi-10T for Vg=4.75 MeV (solid line), black and 0.495V (dashed line, net), (b) Vg=4.75 MeV for Bi-5T (solid black line, left and bottom axes) and -10T (dashed red line, right and top axes). J=0.5meV.

Fig. 4. (a) IV characteristics in meta connected ring for (a) terminal current and (b) circular current at Bi-5T. The inset in (a) shows spin resolved density of states at 10 eV bias.

Fig. 5. (a) Spin polarization and (b) Spin filter efficiency for Bi-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-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.

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