Electrical and Thermal Control of Magnetic Exchange Interactions

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We investigate the far-from-equilibrium nature of magnetic anisotropy and exchange interactions between molecular magnets embedded in a tunnel junction. By mapping to an effective spin model, these magnetic interactions can be divided into three types: isotropic Heisenberg, anisotropic Ising, and anisotropic Dzyaloshinski-Moriya contributions, which are attributed to the background nonequilibrium electronic structures. We further demonstrate that both the magnetic self and exchange interactions can be controlled either electrically by gating and tuning voltage bias, or thermally by adjusting temperature bias. We show that the Heisenberg and Ising interactions scale linearly, while the Dzyaloshinski-Moriya interaction scales quadratically, with the molecule-lead coupling strength. The interactions scale linearly with the effective spin-polarizations of the leads and the molecular coherence. Our results pave a way for smart control of magnetic exchange interactions at atomic and molecular levels.

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Magnetic interactions is a field of continuous intense activities addressing questions ranging from fundamental physics to technological applications. While control of magnetic interactions is straightforward using magnetic fields, control by means of electric field presently is an emerging technique. Technological advances such as magnetic memories, magnetic logic gates, and quantum computation, can be envisioned once current controlled magnetic logic circuits have been achieved.

On the one hand, as the technological advances are striving towards the atomic and molecular scale, experiments on magnetic atoms adsorbed onto different surface materials have demonstrated anisotropic effects on spin excitations [1–4], anisotropic Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction [5], entanglement of spin excitations and Kondo effect [6–8], and formation of stable magnetic configurations [9–11]. Molecular magnets have also been realized in various molecular complexes comprising transition metal atoms [12–18], single molecular magnets [19, 20] and anti-ferromagnetic rings [21–27]. These experimental advances open new alternatives to design multi-functionalities of nanoscale devices [23, 28–32].

On the other hand, the theoretical understanding of magnetic interactions at nanoscale develops fast. Recent theoretical advances include phenomenological and microscopic descriptions of spin dynamics [33, 34], non-equilibrium formulation of RKKY interaction [35], detailed analysis of exchange interactions in non-collinear magnetic materials [36], and magnetic anisotropy in quantum spintronics [37]. However, a comprehensive fundamental understanding of the microscopic mechanism of magnetic interactions is still lacking, which hinders us from more flexible control of spin dynamics at nanoscale.

Here, we uncover the far-from-equilibrium nature of magnetic interactions between molecular magnets embedded between metallic leads. We find that magnetic self and exchange interactions, which are effectively mediated by the electrons flowing between the leads, can be partitioned into isotropic Heisenberg, anisotropic Ising, and anisotropic Dzyaloshinski-Moriya (DM) interactions. The first two interactions scale linearly with the coupling strength to the leads while the DM interaction scales quadratically with the coupling strength. The interactions, moreover, scale linearly with the effective spin-polarizations of the leads and the molecular coherence. We demonstrate that both the magnitude and the character of the interaction, i.e. ferromagnetic or anti-ferromagnetic, can be controlled electrically by gating and tuning voltage bias, and thermally by adjusting temperature bias between the leads. Our results for the self interactions reproduce and generalize the results for magnetic anisotropy discussed in Ref. 37, hence our focus in this paper is on the exchange interactions.

We model the magnetic molecule $n$ by a spin moment $S_n$, which is coupled to a single level via exchange, see Fig. 1. The molecules are coupled to one another through

![FIG. 1: (Color online) Sketch of magnetic molecules embedded in a junction between magnetic leads. Electrons may tunnel between the electrodes and the localized levels $\varepsilon_n$ and between the levels. An electron residing in level $n$ interacts with the localized spin moment $S_n$.](image-url)
tunneling, with rate $T_{\chi}$, and to electrodes, with rate $T_{\chi L}$, $\chi = L, R$, where $L$ ($R$) denotes the left (right) lead. The leads are specified by their respective chemical potential $\mu_\chi$ and temperature $T_\chi$. Thereafter, we consider constant voltage and thermal bias. We can use a Hamiltonian of the following type to represent the interactions in the system:

$$H = H_L + H_R + H_T + H_M + H_{\text{int}}.$$  \hfill (1)

Here, $H_\chi = \sum_{k\sigma}(\epsilon_{k\sigma} - \mu_\chi) c_{k\sigma}^\dagger c_{k\sigma}$ represents the Hamiltonian for the lead $\chi$ and we shall use $p(q)$ for the left (right) lead. The tunneling Hamiltonian $H_T = H_{TL} + H_{TR}$, where $H_{TL} = T_L \sum_{p\sigma} c_{p\sigma}^\dagger c_{p\sigma} + H.c.$, and analogously for the right interaction. We will assume that the spin is conserved in the tunneling process. Further, the molecular complex is represented by $H_M = \sum_{n\sigma}(\epsilon_n \sigma^\dagger \sigma_n e] + T_{\chi}(c_{n\sigma} c_{n\sigma}^\dagger + H.c.)$. The interaction between the de-localized and localized molecular spin $s_n$ and $S_n$, respectively, is written as $H_{\text{int}} = \sum_n J_n s_n \cdot S_n$, where $s_n = \sum_{\sigma\sigma'} c_{n\sigma}^\dagger \sigma_{n\sigma'} \sigma_{n\sigma'}/2$, whereas $\sigma$ is the vector of Pauli matrices [38].

The local interactions between the spin moment $S_n$ and electrons in level $\epsilon_n$ give rise to a contribution $\delta S$ to effective spin action $S_{\text{eff}}$ [34, 39, 40], given by

$$\delta S = \frac{1}{\epsilon} \sum_n \int \left[ \epsilon_m j_{mn}(t, t') + S_m(t) \cdot j_{mn}(t, t') \right] S_n(t) dt dt', \hfill (2)$$

The contribution $\delta S$ for $j_{mn}$ provides the magnetic field exerted on the local spin magnet due to electron flow. Here, $\epsilon_m = \text{diag}[\epsilon_m, \epsilon_m]$ and $s_m^{(0)}(t) = \sum_{\sigma\sigma'} c_{m\sigma}^\dagger \sigma_{m\sigma'}$ is the charge, where $\sigma_0$ is the identity matrix. The current $j_{mn} = i\epsilon_m j_{mn}\theta(t - t')(s_m^{(0)}(t), s_n(t'))$ carry the magnetic anisotropy and exchange interactions between the local magnetic moments $S_n$. As the first contribution in Eq. (2) was discussed in [37], our primary focus will be on the second.

The self interaction $j_{mm}$ defines the anisotropy field acting on the local spin moment $S_m$, while $j_{nm}$ mediate the exchange interaction between two different spin moment $S_n$ and $S_m$. For small coupling $j_m$ we can neglect the back-action from the localized spins on the electrons. In the stationary regime we can therefore express the current $j_{mn}$ in energy space as

$$j_{mn}(\epsilon) = \frac{e}{4j_m} \int_{-\epsilon}^{\epsilon} \frac{1}{\omega - \epsilon + i\delta} d\omega \text{sp} \left( \sigma G_{mn}(\epsilon) \sigma G_{nm}^*(\epsilon') \right) \frac{d\epsilon'}{2\pi} \frac{d\epsilon}{2\pi}. \hfill (3)$$

Here, $G_{\sigma^/>}$ is the lesser/greater (spin matrix) Green function (GF) for propagation of an electron from molecule $m$ to $n$. $\text{sp}[\sigma G_{mn}]$ are dyads defined as $a b = a_i b_i \hat{j}$ such that $j_{mn}$ constitutes a tensorial quantity.

The electron GF $G_{mn}$ can always be partitioned into charge and magnetic components, $G_{mn}^{(0)}$ and $G_{mn}^{(1)}$, according to $G_{mn} = G_{mn}^{(0)} + G_{mn}^{(1)} \sigma$. In terms of this notation it is straightforward to see that the local molecular spins in Eq. (1) can be mapped into an effective Hamiltonian $H_S$ corresponding to the interaction $S_m \cdot S_n + D_{mn} \times S_n$, given by

$$H_S = -\sum_{mn} S_m \cdot (J_m S_n + I_m \cdot S_n + D_{mn} \times S_n), \hfill (4)$$

where the three contributions in the above model describe Heisenberg, Ising, and DM interactions, respectively.

$$I_{mn}(\alpha) = \frac{1}{2} j_m j_n \int \frac{1}{\omega - \epsilon + i\delta} \left( G_{mn}(\epsilon) G_{nm}(\epsilon') - G_{mn}^{(0)}(\epsilon) G_{nm}(\epsilon') - G_{mn}(\epsilon) G_{nm}^{(0)}(\epsilon') \right) d\epsilon \frac{d\epsilon'}{2\pi}, \hfill (5a)$$

$$D_{mn}(\alpha) = \frac{1}{4} j_m j_n \int \frac{1}{\omega - \epsilon + i\delta} \left( G_{mn}^{(0)}(\epsilon + \alpha) G_{nm}^{(1)}(\epsilon) - G_{mn}^{(1)}(\epsilon + \alpha) G_{nm}^{(0)}(\epsilon) \right) d\epsilon \frac{d\epsilon'}{2\pi}, \hfill (5b)$$

$$G_{mn}^{(0)}(\epsilon + \alpha) G_{nm}^{(1)}(\epsilon) - G_{mn}^{(1)}(\epsilon + \alpha) G_{nm}^{(0)}(\epsilon) \hfill (5c)$$

in the limit $\alpha \to 0$ ($\hat{\alpha}$ denotes the Cauchy principal value). Negative (positive) parameters $j_m, j_n$, and $D_{mn}$ correspond to ferromagnetic (anti-ferromagnetic) interactions.

We notice here, for instance, that the Heisenberg like interaction is finite regardless of the spin-polarization in the molecules, while the Ising and DM like interactions are finite only under spin-polarized conditions. It may also be noticed that the Ising like interaction contributes to the uniaxial anisotropy [41] whereas the DM like interaction provides a transverse anisotropy component.

The expressions for the Heisenberg, Ising, and DM self and exchange interactions given in Eq. (5) constitute a very general result since they provide the spin-interactions far from equilibrium, as well as in equilibrium, both under electric and thermal fields. The expressions can, moreover, be employed in materials calculations by interpreting the GFs $G_{mn}$ in terms of real space distributions of the electronic structure. In the present context, we shall go deeper into a discussion of their properties in coupled magnetic molecules.

Under equilibrium conditions (vanishing voltage and thermal biases), we can employ the fluctuation-dissipation theorem through the relation $G_{mn}^{(1)}(\alpha) = \cdots$
(±i)\(f(\pm\omega)|−2\text{Im}G_{mn}(\omega)|\), where \(f(\omega)|\) is the Fermi function at the (electro-) chemical potential \(\mu\). We define \(\Delta_{mn}^{(0)} = \sum c_{m}G_{mn}(\omega)/2\) and \(\Delta_{mn}^{(F)} = \bar{2}\sum c_{m}G_{mn}(\omega)/2\), for a simple collinear spin-polarized structure. Inserting into Eq. (5a) and using Kramers-Kröning’s relations we obtain
\[J_{mn} = j_{m} = \sum_{c}G'_{mn}(\omega)\chi_{\sigma\sigma}(\omega)/\omega - E_{as}.\]

Here, \(E_{\sigma\pm} = (\epsilon_{\sigma\pm} + \epsilon_{2\sigma\pm} + i\epsilon_{\sigma\pm} - i\Gamma_{\sigma\pm}/2)/2, \Omega^{\pm} = (\Lambda_{\sigma\pm} - i\Gamma_{\sigma\pm}/2)^{2} + 4\Delta^{2}, \Lambda_{\sigma\pm} = \epsilon_{\sigma\pm} - i\Gamma_{\sigma\pm}, \sigma_{\sigma} = \sum_{\chi}n_{\chi}^{0}, \) and \(\gamma'_{\sigma\pm} = \Gamma_{\sigma\pm}^{2} - \Gamma_{\sigma\pm}^{2}.\) The resonance \(E_{\sigma\pm}(E_{\sigma\mp})\) signifies the orbital with the highest (lowest) energy, and \(G_{\chi\chi}^{0} = 2\pi\sum_{k\chi}T_{\chi\chi}^{k}\rho_{k\chi}^{0}\) denotes the coupling to the lead \(\chi = L,R\), in terms of the density of electron states (DOS) \(\rho_{k\chi}^{0}\). The spin-polarization in the leads is parametrized within a Stoner picture using \(\delta_{\chi\chi} = L,R\), in terms of the density of electron states (DOS) \(\rho_{k\chi}^{0}\).

For transparency of the mathematical formulation, we assume equivalent molecules such that \(\epsilon_{\sigma\sigma} = \epsilon_{0}\) and symmetric couplings \(\Gamma_{\sigma}^{0} = \Gamma_{\sigma}/2\), retaining spin-polarization in the leads. The Heisenberg exchange \(J_{mn}(m \neq n)\) then becomes
\[J_{mn} = -T^{2}c/8\pi j_{m} = j_{m} = \sum_{c}G'_{mn}(\omega)\chi_{\sigma\sigma}(\omega)/\omega - E_{as}.\]

We notice that the Heisenberg exchange depends on the electronic occupations (\(\propto f_{\sigma} + f_{\bar{\sigma}}\)) of the leads and scales linearly with \(\Gamma\). The expression, moreover, indicates that there is a finite exchange interaction between the localized spins whenever the chemical potential \(\mu\) lies within the energy range of the molecular orbitals, that is, \((\mu - \epsilon_{0})^{2} \leq \Delta^{2} + (\Gamma_{\sigma}/4)^{2}\). This result is demonstrated in Fig. 2(a), which shows the equilibrium exchange as function of \(\mu = \mu\) for different spin-polarizations \(p_{L}, p_{R}\). The exchange, which peaks at the orbital resonances \(E_{asl}\), is anti-ferromagnetic below \(E_{asl}\) (above \(E_{asl}\)) and ferromagnetic between the resonances, which is a typical behavior for superexchange. This behavior can be controlled by means of gating or tuning voltage bias, see Fig. 2(b) where the system is gated \((\mu - \epsilon_{0} = -2)\) and driven with a finite voltage bias.

From Fig. 2(a) and 2(b), it is clear that the equilibrium and non-equilibrium responses on the spin-polarization in the leads are quite different. While the exchange depends only weakly on \((p_{L}, p_{R})\) in equilibrium, the ferromagnetic regimes changes dramatically under non-equilibrium conditions. Current flowing from stronger to weaker spin-polarization generates a stronger ferromagnetic exchange while it becomes weaker when the current flows in the opposite direction.

Varying the temperature and/or introducing a thermal bias \(\Delta T = T_{R} - T_{L}\) provides an alternative route to control the exchange. The thermal broadening of the electronic density in the leads effectively makes it (partially) resonant with the molecular orbitals. The plots in Fig. 2(c) shows the dependence on a thermal bias for different \((p_{L}, p_{R})\). The initial peak corresponds to that the lower orbital, c.f. Fig. 2(a) and 2(b), becomes resonant with the thermally broadened electrons in the right lead. With increasing \(\Delta T\), more of the molecular electron density contributes to the process, balancing ferromagnetic and anti-ferromagnetic exchange, which results in a decreased total exchange interaction. The plots in Fig. 2(c) shows that we can control this balance into a regime of ferromagnetic exchange for a finite range of temperature biases by tuning the spin-polarizations in the leads.

Although previous studies uncovered that the sign of Heisenberg exchange interaction among magnetic impurities can be tuned electrically (see, e.g., [45, 46]), to our knowledge this thermal control of the Heisenberg exchange has never been explored before. More importantly, our general results Eqs. (4), (5), provide a unified microscopic theory for both the electrical and thermal control of magnetic interactions including also anisotropic interactions, as we discuss below.

Under the same conditions as above, we write the Ising exchange \(I_{mn} = I_{mn}2\hat{z} (m \neq n)\) where

\[I_{mn} = -T^{2}c/4\pi j_{m} = j_{m} = \sum_{\sigma\sigma}G_{\sigma\sigma}(\omega)\chi_{\sigma\sigma}(\omega)/\omega - E_{as}.\]

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\[I_{mn} = -T^{2}c/4\pi j_{m} = j_{m} = \sum_{\sigma\sigma}G_{\sigma\sigma}(\omega)\chi_{\sigma\sigma}(\omega)/\omega - E_{as}.\]

The basic difference compared to the Heisenberg ex-
change is that the Ising exchange requires a non-vanishing spin-polarization in the system to be finite. Effectively, the Ising energy becomes a measure of the spin-polarization in the system, which is indicated by the presence of the z-component of the Pauli matrices in Eq. (8). Therefore, the Ising energy is small everywhere except when the molecular orbitals are resonant with the chemical potential(s) of the lead(s), see Fig. 3. In a similar way as with the Heisenberg energy, we can tune the sign of the Ising exchange by means of gating, voltage bias, thermal bias, and spin-polarization.

We finally write the DM exchange energy $D_{mn} = D_{mn} \hat{2}$ ($m \neq n$) within the same approximation but let $p_L$ and $p_R$ be independent. For this we obtain

$$D_{mn} = -\frac{1}{16 \pi} \int \Re \int \frac{1}{|\Gamma_L \Gamma_R|^2} \left( \Gamma_L^\dagger \Gamma_R^\dagger - \Gamma_L \Gamma_R \right) \int \left( f_L(\epsilon) - f_R(\epsilon) \right) \times \frac{(\epsilon - \epsilon_0)^2}{|\epsilon - E_{1\uparrow}|^2|\epsilon - E_{1\downarrow}|^2|\epsilon - E_{2\downarrow}|^2|\epsilon - E_{2\uparrow}|^2} d\epsilon. \quad (9)$$

The integrand peaks at the resonances $E_{1\sigma}$ while the sign of $D_{mn}$ is governed by the polarities of the voltage bias and temperature difference, and the spin-polarization in the leads. This expression also shows that the DM energy results from breaking the time-reversal symmetry (spin-polarized current between the localized spins) and space inversion symmetry (biased by a source-drain voltage and/or temperature difference), see Fig. 4. The scaling with $\Gamma^2$ suggests that the influence of $D_{mn}$ on the spin excitation spectrum becomes important for stronger coupling $\Gamma$. The combination $\Gamma_L^\dagger \Gamma_L - \Gamma_R^\dagger \Gamma_R$, which corresponds to an effective spin-orbit coupling between the leads, suggests that $D_{mn}$ is maximal for antiferromagnetic alignment.

For small voltage bias and zero temperature difference, we have $f_L(\epsilon) - f_R(\epsilon) \approx eV(\beta/4)\cosh^{-2}\beta(\epsilon - \mu)/2$, which indicates a linear voltage bias dependence of $D_{mn}$ near equilibrium, as is shown in Fig. 4(b). In case of small temperature difference $\Delta T = T_R - T_L$ and vanishing voltage bias, we have $f_L(\epsilon) - f_R(\epsilon) \approx -2(\Delta T/T)\beta/4)(\epsilon - \mu)\cosh^{-2}\beta(\epsilon - \mu)/2$, indicating a linear dependence on the temperature difference, see Fig. 4(c).

The conclusions from our present study of the electrically and thermally mediated exchange interactions between localized magnetic moments have an impact on the magnetic properties of magnetically active quantum devices designed with atomic or molecular building blocks. Depending not only on the couplings to the leads and the spin-polarization in the system but also on gating, voltage bias, and effective temperature difference between the leads, the expected magnetic properties may be drastically different. We expect that our findings should be experimentally verifiable by means of existing state-of-the-art. We believe that our presented results provide essential new understanding to magnetic interactions and the ability for control by means of external electric and thermal sources.

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