Coulomb blockade-tuned indirect exchange in ferromagnetic nanostructures

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Abstract

We develop a theory of the reversible switching of the magnetic state of the ferromagnet-insulator-normal metal-ferromagnet (FINF) nanostructure. The switching is controlled by tuning the Coulomb blockade strength via the gate voltage on the normal metal granule. The proposed mechanism allows for realizing the switching without passing a dissipative current through the structure.

Keywords: Ferromagnet hybrid structures, spin valve, Coulomb blockade, spin current

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1. Introduction

The pioneering work by Slonczewskii [1] who proposed switching of the magnetic state in the Ferromagnet/Normal metal/Insulator/Ferromagnet (FNIF) multilayer structure by applying a non-equilibrium spin current, opened a novel exciting direction in the study of magnetic heterostructures with tunable configurations. Following the concept of [1], subsequent papers [2, 3] suggested an interesting possibility of rotating magnetization in magnetic bilayer structures containing an insulating layer (FIF structure) by the applied bias. It was shown that in a tunneling bilayer, the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between the magnetic moments in ferromagnetic layers would oscillate as a function of the bias. This allows for tuning the sign of the interaction alternating between the ferromagnetic and antiferromagnetic coupling by tuning the bias. Devices with the bias-controlled switching magnetization are advantageous as compared to those controlled by the external magnetic field, since the latter is hard to localize within the small (several nanometers size) area. On the other hand, the bias results in the current through a device leading to dissipation and, thus, to power losses.

In this work, building on the ideas of Refs. [2] and [3] we investigate the effect of the nanoscale tunnelling through the hybrid F1-I-N-F2 structure in which one of the ferromagnetic islands, F2, see Fig. 1, is small enough for the Coulomb blockade effects become essential. We show that in such a structure the change of the sign of RKKY coupling can be achieved by tuning the electron polarization by the bias without any current passing across the system.
2. The Model

Let us consider a multilayered system with the structure Fe1-I-N-Fe2 (Fig. 1) where Fe1 is a bulk ferromagnet, I and N mark insulating and normal layers, respectively, and Fe2 is a ferromagnetic layer shaped as a small metal granule. In what follows we will consider an indirect exchange between the Fe1 and Fe2 following the simple model of [4]. Importantly, the indirect exchange implies the coupling between magnetic ions localized in different ferromagnets. The coupling is thus mediated by the conducting electrons whose paths traverse between the two ferromagnets. Summation over different magnetic ions leads to the exchange interaction between all the spins of the respective ferromagnets, i.e., between their respective magnetizations. Hence the interaction between the layers appears as a sum of the pair interactions between the individual spins belonging in different ferromagnets. In turn, the interactions between the individual spins are
described by the second order perturbation theory with respect to spin coupling to the propagating electron modes. The interface between the ferromagnet and the normal metal is assumed to be perfect, and the metal is supposed to have a simple band structure. This model captures essential features of the non-dissipative magnetization switch, and without any loss of generality, we can omit the details of the interlayer coupling such as surface imperfections, quantum well states, see, e.g., [5–7].

The Hamiltonian describing the propagating electron modes \( \alpha \) reads

\[
H = \sum_\alpha \varepsilon_{\alpha,b} a_{\alpha,b}^+ a_{\alpha,b} + \sum_\alpha \varepsilon_{\alpha,g} a_{\alpha,g}^+ a_{\alpha,g} + \sum_\alpha \left( A_T a_{\alpha,b}^+ a_{\alpha,g} + \text{h. c.} \right) + J \sum_{j,\alpha} S_j s_{e,\alpha} a_{\alpha,b}^+ a_{\alpha,b} + J \sum_{i,\alpha} S_i s_{e,\alpha} a_{\alpha,g}^+ a_{\alpha,g}.
\]

(1)

Here \( a_{\alpha,b}^+, a_{\alpha,g}^+ \) are creation operators for the mode \( \alpha \) in the bulk and in the granule, respectively and \( a_{\alpha,b} \) and \( a_{\alpha,b} \) are the corresponding annihilation operators, \( A_T \) is the tunneling amplitude, \( s_e \) is the electron spin, \( S_j, S_i \) are localized spins within the bulk and in the granule, respectively, and \( J \) is the exchange integral.

Before moving further, it is instructive to compare our scheme with the previous approaches dealing with the electron transport in nano-aggregates including magnetic elements. The paper [8] reports theoretical studies of Kondo effect in quantum dots between the two ferromagnetic layers. The discussed phenomena are related to the correlations between the electrons within the ferromagnetic leads and within (non-ferromagnetic) granule, which finally causes an influence of these leads on the spin state of the granule. At variance, we describe the electron-mediated coupling between the ferromagnetic ions located within the granule and in a single ferromagnetic layer. This coupling has a form of in the effective exchange interaction between the total spins of the layer and that of the granule,
i.e., between their magnetizations. Note, further, that while the approach by [8] accounts only for the electron-electron interactions, the effect we report here is based on a subtle interplay between the charging energy of the granule and the energy dependence of the electronic states mediating the indirect exchange between the ferromagnetic layer and the granule. The energy dependence of the electronic states, in turn, transforms into the gate voltage dependence. As a result, the indirect exchange resulting from the interference of different electron trajectories coupling the two magnetic ions becomes dependent on the gate voltage while the Coulomb blockade effect impedes the current through the structure even in the presence of the gate voltage. Note further that the design of the device of [9] is critically different from ours shown in Fig. 1. The latter consists of the ferromagnetic layer and ferromagnetic granule separated by the passive interlayer including the insulating and the normal-metal parts respectively, the latter being in perfect contact with the granule.

3. Calculations and results

Starting with the Hamiltonian given by Eq. (1), we calculate the interaction energy for the pair of localized spins $i, j$ considering it as the result of coupling spin $i$ to the Friedel oscillations of the spin density of delocalized electrons produced by spin $j$ [10], see Fig 2. Consequently, the Friedel oscillations arise from the interference between the unperturbed electron mode and the scattering wave produced by spin $j$.

Notice first, that the tunneling coupling hybridizes the states in the bulk with the states within the granule. Thus, the propagating mode $|\alpha; b\rangle$ of the bulk acquires, in the first approximation with respect to $A_T$, an admixture $|\alpha; g\rangle$ spreading
Figure 2: A sketch of a general indirect exchange between the localized spins $j$ and $i$ mediated by the delocalized electrons. An incident plain wave $k$ is scattered by the spin-dependent potential of the ion $j$. The interference of the incoming wave with the scattered wave at the location of the spin $i$ produces the local fluctuation of the spin density which is coupled with spin $i$ within the granule and vice versa. The boundary condition at the tunnel barrier is given as $|\alpha; b\rangle = A_T|\alpha; g\rangle$. Similar considerations hold for the states $|\alpha; b, j\rangle$ resulting from scattering of the modes $|\alpha, b\rangle$ by the spin located at the site $j$ within the bulk. Next, we take into account that the state $|\alpha, g\rangle$ can suffer backscattering within the granule at the site $i$ forming the scattering state $|\alpha; g, i\rangle$. In turn, in the second order with respect to the tunneling amplitude, it creates an addition $|\alpha; gran, i; bulk\rangle$ within the bulk layer. This processes are shown in Fig. 3. As a result, the propagating state resulting from the initial mode $\alpha$ assumes the form

$$|\alpha\rangle = |\alpha; b\rangle + |\alpha; g\rangle + \sum_j (|\alpha; b, j\rangle + |\alpha; b, j; g\rangle) + \sum_i (|\alpha; g, i\rangle + |\alpha; g, i; b\rangle).$$ (2)

The similar procedure applies to the states $\psi_\beta$ which for vanishing tunnel transparency are localized within the grain. Then any contribution of the mode $\alpha$ to the interaction energy acquires the form $\langle \alpha; g | S | \alpha; b, j; g \rangle$. The resulting interaction energy is obtained by the summation over the modes $\alpha$ and $\beta$. As a result, in the
Figure 3: A sketch of the processes leading to the indirect exchange between the ions $j$ and $i$ located at the interfaces of ferromagnetic layers F1 and F2, respectively, mediated by the incident wave from the layer F1 mode $\alpha$ with the wave vector $\vec{k}$. (a) The interference between the incident mode $\alpha$, propagating through the tunnel junction and the normal layer to the ferromagnetic layer F2, and the result of scattering of the mode $\alpha$ by the ion $j$ at the point of location of the ion $i$. The index $t$ denotes the transmitted part of the wave confined within the normal layer, indices $s(i, j)$ describe scattering of the mode by the respective ion. The exchange occurs due to modulation of the spin density of the delocalized modes at the locations of the ions $i, j$. This modulation results from the interference between the two different paths involving both tunneling- and scattering events, respectively. The dependence of the wave vector $\vec{k}(V)$ on the gate voltage allows for controlling the phase of the interference, and, thus, controlling the sign of the exchange, by the gate voltage $V$. (b) The interference between the incident mode $\alpha$, and the mode, resulting from its scattering by the ion $i$, and then propagating in the opposite direction through the normal layer and the tunnel junction. The interference contribution is accounted at the position of the ion $j$.

In the lowest approximation in the tunnel transparency, the indirect exchange coupling between the two ferromagnetic ions located in the different ferromagnetic layers...
\[ U_{\text{ex}}(i, j) \simeq J a^3 \sum_{\alpha, \beta, s_e} [S_i s_e \psi_{\alpha; g}(R_i) \psi_{\alpha; j, i; g}^*(R_j) + S_j s_e \psi_{\alpha; h}(R_j) \psi_{\alpha; i, j; h}^*(R_i)] + S_j s_e \psi_{\beta; g, i; h}(R_j) + S_i s_e \psi_{\beta; g, j; i}(R_i) + \text{c. c.}] \quad (3) \]

Here \( J \) is the local exchange integral magnitude, \( a \) is the size of elementary cell, \( s_e \) is the electron spin while \( \text{c. c.} \) means complex conjunction. The first two terms describe the interference of the electron states at sites \( i \) and \( j \), respectively, while the two last terms have the similar nature, but stem from modes \( \beta \) localized within the granule. Spin dependences of the scattering amplitudes \( \psi_{\alpha; i, j; g} \) are shown later, Eq. (6).

One notes that the admixtures to the states \( \alpha \) resulting from the hybridization within the granule are formed from the states \( \beta \) having the same energy as modes \( \alpha \). At the same time, the tunneling of an electron from the bulk to the granule increases the Coulomb energy of the granule. If the gate creates a voltage \( V \) between the granule and the bulk, an electron having the kinetic energy \( \varepsilon \) in the bulk, acquires the kinetic energy

\[ \varepsilon' = \varepsilon - eV - e^2 / C \quad (4) \]

upon tunneling into the granule, where \( C \) is the granule capacitance. When a hole tunnels, the state with the energy \( \varepsilon \) of the grain is coupled to the state with the energy

\[ \varepsilon' = \varepsilon - eV + e^2 / C \quad (5) \]

in the bulk, see Fig. 3. One has to bear in mind that the tunneling of a hole from the bulk to the granule corresponds to the tunneling of an electron from the granule to the bulk and vice versa. One also notes that the terms in Eq. (3) involving modes
\(\alpha\) describe tunneling of an electron to the granule while the modes \(\beta\) describe tunneling of an electron from the granule. One has to note further that for \(|eV| < e^2/C\), the real tunneling processes are suppressed by the Coulomb blockade and the effects become virtual. Note, however, that the relations between the energies of the states hybridized by tunneling given above hold irrespective to the fact whether tunneling is real or virtual since these relations do not depend on the occupation numbers of the relevant states. It is the situation of virtual tunneling allowing eliminating direct current through the structure that will be considered in what follows.

We chose the size of the granule to be much larger than the electron wavelength, this implies that the states with a given wave vector can be considered as the basis. Note that while one could have been expecting that \(\alpha\)-modes correspond to \(k_x > 0\) and the \(\beta\)-modes correspond to \(k_x < 0\), this appears not be the case. Indeed, let us consider an electron mode with \(k_x < 0\) incident from the granule to the interface with the tunneling layer. The mode is partly reflected back to the granule acquiring \(k_x > 0\) and partly tunnels to the bank. The following interference schemes can be realized: (i) The wave scattered from the ion \(j\) in the bulk tunnels back to the granule and interferes on the ion \(i\) with the wave reflected from the interface, (ii) the reflected wave is scattered by the ion \(i\) and the scattered wave tunnels to the bank where it interferes on the ion \(j\) with the tunneling tail of the initial incident mode. One notes that the corresponding interference terms coincide with the ones in Eq. (3) which result from the modes \(\alpha\) with \(k_x > 0\) incident from the bulk. The same considerations can be applied to the modes \(\alpha\) which acquire the terms with \(k_x < 0\) due to acts of reflection from the back boundary of the granule. These additional terms in Eq. (3) are included as complex conjunction of
the first four terms.

The propagating exponentials entering \( \psi_{\alpha}, \psi_{\beta} \) have a form \( \psi = a^{-3/2} \exp(i k R) \) where we have used a normalization with respect to the volume of an elementary cell \( a^3 \). The scattering amplitudes, \( \psi_{\alpha;b,j,g} \), in the Born approximation are given by

\[
\psi_{\alpha;b,j,g}(R_i, R_j) = \frac{A_T J S_j s_e ma^3}{2\pi \hbar^2 |R_j - R_i| a^3 \exp \left[ i(kR_j + k|R_j - R_i|) \right]. \tag{6}
\]

For other scattering amplitudes one has similar expressions differing by the notations of the scattering ions and by the signs of the wave vectors. Finally inserting the corresponding amplitudes to Eq. (3) after the summation over the electron spins one obtains for the exchange coupling the expression \( U_{\text{ex}}(\tilde{R}_i - \tilde{R}_j) \) where

\[
U_{\text{ex}}(\tilde{R}) = \frac{J^2 S_i S_j m a^3 |A_T|^2}{2\pi \hbar^2 \tilde{R}} \times \sum_{k,\nu} \left( 2e^{i(k\nu_{\nu',\tilde{R}} - k'_{\nu',\tilde{R}})} + e^{i(-k\nu_{\nu',\tilde{R}} - k'_{\nu',\tilde{R}})} + e^{i(-k\nu_{\nu',\tilde{R}} + k'_{\nu',\tilde{R}})} \right) \tag{7}
\]

where \( k' \) is the wave vector for the state with the kinetic energy \( \varepsilon' \) (thus depending on the gate voltage). Here we redefined the spatial coordinates, \( \tilde{R} \), extracting the thickness of the insulating layer, \( \tilde{R} \equiv |\tilde{R}| \). Indices \( \nu = 1, 2 \) stand for the electron and hole channels, respectively. In general, the factor \( |A_T|^2 \) depends on the electron wave vector \( k \). However this dependence is relatively weak as compared to that resulting from the strongly oscillating exponential and does not influence the effect of the sign change. Thus, in the lowest approximation one can neglect the dependence of \( A_T \) on the \( k \) and the applied voltage.

Let us first perform an integration over directions of \( k'_{\nu} \) which enter through the combinations \( \exp(i k'_{\nu,\tilde{R}} \cos \theta) \), where \( \theta \) is an angle between the vectors \( k'_{\nu} \) and \( \tilde{R} \). The integration over \( \theta \) of the exponential given above yields \( (2/k'_{\nu}) \sin(k'_{\nu} \tilde{R}) \)
Performing similar integration of all the terms entering r.h.s. of Eq. (7) we obtain

\[ U_{\text{ex}}(\tilde{R}_{ij}) = J^2 S_i S_j \frac{ma^3 |A_T|^2}{\pi \hbar^2 R_{ij}^3} \sum_{k',\nu} \sin \frac{2k'_\nu \tilde{R}_{ij}}{k'_\nu} \sin k_{ij}' \nu \]  

(8)

Now let us perform an integration over \( k \). Based on the relations (4) and (5) between the energies of electronic states hybridized by tunneling we have:

\[ k'_1 = k \left[ 1 - \frac{1}{2} \left( \frac{eV + e^2/C}{\varepsilon} \right) \right], \quad k'_2 = k \left[ 1 - \frac{1}{2} \left( \frac{eV - e^2/C}{\varepsilon} \right) \right]. \]

(9)

It is important that an electron can tunnel only \textit{from} an occupied state while the hole can tunnel only \textit{to} an occupied state. Accordingly, for \( \nu = 1 \) the integration over \( k \) is up to \( k_F \), while for \( \nu = 2 \) integration goes up to

\[ k_h = k_F \left( 1 + \frac{eV - e^2/C}{2\varepsilon} \right). \]

(10)

Correspondingly, we have

\[ U_{\text{ex}}(\tilde{R}_{ij}) = J^2 S_i S_j \frac{2ma^3 |A_T|^2}{2\pi \hbar^2 k_F^3 R_{ij}^3} \left\{ \cos \left[ 2k_F \left( 1 - \frac{eV + e^2/C}{2\varepsilon} \right) \tilde{R}_{ij} \right] + \cos(2k_F \tilde{R}_{ij}) \right\}. \]

(11)

Note that the dependence on the gate voltage exists only in the electron channel describing an electron tunneling to the granule and is absent in the hole channel where an electron is tunneling \textit{from} the granule and thus its momentum within the layer of a normal metal is not affected by the gate voltage since this layer is located \textit{within the granule itself}.

Next, making use of Eq. (11) we employ the calculation procedure similar to that of [4]. Namely, we assume that our granule is a slab including intermediate normal layer and separated from the bulk ferromagnet by a tunnel barrier (the latter is a new feature as compared to the model of Ref. [4]). We replace the
summation over ion \( j \) by integration over corresponding spatial coordinates and take into account that the integrand strongly oscillates with the spatial scale of the order of an elementary cell size \( a \). As a result, only the interface ions of the ferromagnet contribute efficiently. Thus only spins \( i \) corresponding to the interface of the ferromagnet are important. Thus we perform an integration over the volume of Fe2 (granule) for the each ion \( i \) at the interface of the Fe1 with the area \( A \).

Note that the most efficient coupling could be expected for the case where the oscillations of the indirect exchange do not cancel each other, that is for the granule of an atomic size. However, at present, the working body of realistic devices on the base of ferromagnetic metals are by far exceeding the atomic dimensions.

Finally, we arrive at the following expression for the coupling between the particles per unit area:

\[
\frac{U_{\text{ex}}}{A} = \frac{J^2 m S_y S_b}{16\pi^2\hbar^2} K(k_Fd),
\]

where \( d \) is a distance between the ferromagnetic layers, \( A \) is the contact area, \( S_y \) and \( S_b \) are the values of localized spins within the bulk and within the grain, respectively, and

\[
K(z) = B \frac{|A_T|^2}{z^2} \left\{ \sin z + \sin \left[ z \left( 1 - \frac{eV + e^2/C}{2\varepsilon_F} \right) \right] \right\},
\]

with \( z = 2k_Fd \), \( B \) being the numerical factor of the order of unity, and \( d \) being a thickness of the normal metal layer. In the course of our derivation we exploited the fact that \( k_F \simeq \pi/a \). For small \( eV \) one finds

\[
K(z) \simeq \frac{|A_T|^2}{z^2} \left\{ \sin z + \sin \left[ z \left( 1 - \frac{e^2}{2C\varepsilon_F} \right) \right] - \frac{eV}{2\varepsilon_F} \cos \left[ z \left( 1 - \frac{e^2}{2C\varepsilon_F} \right) \right] \right\}.
\]

Thus, the voltage-dependent part is

\[
K(z) \simeq - \frac{eV|A_T|^2}{2\varepsilon_F z^2} \cos \left[ z \left( 1 - \frac{e^2}{2C\varepsilon_F} \right) \right].
\]
The sign of this contribution to the exchange energy is changed with the sign of the voltage. Note that the effect we describe is completely different from the mechanisms of switching suggested before in [1–3] since it implies no current through the device and therefore, no dissipation.

4. Discussion and estimates

One of the most promising ways for an experimental realization of the proposed structure is the point contact nanofabrication technique (see, e.g., [11–13]). It allows, in particular, manipulating with ferromagnetic or normal metal granules with a size of a few nanometers [11, 12], or even with single molecules [13]. Manipulation with metallic layers with a thickness $\sim 1$ nm within the point contact was reported in [12] while a presence of tunnel barriers [12, 13] can be used to fabricate gated structures. We suggest that the experimental system is to be produced on the base of a ferromagnetic granule covered by a normal metal layer fabricated within the nanoscale point contact between ferromagnetic (Fe1) and normal electrodes, the latter playing a role of the gate. The magnetic state of the granule can be probed by application of short pulses of large bias (lifting the Coulomb blockade) since the current through the structure is sensitive to mutual orientation of magnetization in Fe1 and Fe2.

For a typical size of the grain of 5 nm one estimates $e^2/C \sim 0.1$ eV and $k_F d \sim 10$. Thus the Coulomb blockade regime exists until $V < 0.1$ V while the coupling in this regime according to Eq. (13) oscillates with the variation of $V$.

Let us evaluate the relative efficiency of the bias-controlled exchange with respect to the magnetic dipole interaction. In typical ferromagnets the ratio of the dipole interaction, $E_d \sim \mu^2/a^3$, where $\mu$ is a magnetic moment of a magnetic ion
while $a$ is a lattice constant, to direct exchange energy, $E_{ex}$, for neighboring spins is of the order of $10^{-4}$. The effective magnetic field near the surface of the larger ferromagnet is $H_D \sim \gamma(\mu/a^3)$ where $\gamma$ is demagnetizing factor. For a slab with a thickness $b$ and a linear size $c \gg b$ it is of the order of $b/c \ll 1$. Thus the total dipole energy is of the order of $H_D \mu V/a^3$ where $V$ is a volume of the smaller ferromagnetic particle. If the latter is a slab with a thickness $t$ and a linear size $L$, $V \sim tL^2$. Correspondingly, the dipole energy $E_D \sim \gamma E_d(tL^2/a^3)$.

Making use of Eqs. (11) and (13), one estimates the efficiency of the bias-controlled exchange for $(eV/\varepsilon_F)k_Fd > 1$ as $E_X \sim U_{ex}(L/a)^2|A_T|^2/(k_Fd)^2$. Correspondingly,

$$\frac{E_X}{E_D} \sim \gamma^{-1}\left(\frac{U_{ex}}{E_d}\right)\left(\frac{a^5}{d^2tL^2}\right)^2 \left(\frac{L^2|A_T|^2}{a^2}\right).$$

(15)

The Coulomb blockade conditions imply that $|A_T|^2(L/a)^2 < 1$. As a result, the bias-controlled exchange dominates the dipole interactions provided

$$\gamma^{-1}\left(\frac{U_{ex}}{E_d}\right)\left(\frac{a^5}{d^2tL^2}\right) > 1.$$  

(16)

Taking $t \sim L$, $d \sim 1$ nm one sees that this condition is satisfied if $L \leq 5$ nm provided $\gamma = 10^{-1}$ and is experimentally realistic [11–13].

The switching of the granule magnetization can be, in particular, registered due to Giant Magnetoresistance effect studying a (weak) direct current through the structure. As it is known, the resistance of the spin valve of the sort of studied in our paper is sensitive to mutual orientation of magnetizations of the magnetic layers.

The following note is in order. Our simplified approach does not take into account the surface imperfectness and realistic band structure, and we present only the order of magnitude estimates. Yet, our conclusions that are based on
the facts that (i) the RKKI interaction strongly oscillates as a function of distance between the interacting spins and (ii) the period of the oscillations depends on the actual electron wave vector which can be affected by the applied electric field are not affected by the details the surface and band structures.

5. Summary

We have proposed a theory of the effect of the dissipationless magnetic coupling in a hybrid structure consisting of two ferromagnetic layers separated by a tunnel barrier and normal metal, which utilizes the Coulomb blockade effects, suppressing the real tunneling processes, and realizes the switch controlling the sign of the coupling.

Acknowledgments

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