Magnetic Domain Walls in Double Exchange Materials

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We study magnetic domain walls in double exchange materials. The domain wall width is proportional to the square root of the stiffness. In the double exchange model the stiffness has two terms: the kinetic energy and the Hartree term. The kinetic energy term comes from the decrease of the tunneling amplitude in the domain wall region. The Hartree term appears only in double exchange materials and it comes from the connection between band-width and magnetization. We also calculate the low-field magnetoresistance associated with the existence of magnetic domains. We find a magnetoresistance of 1 – 2%. The magnetoresistance can be considerably larger in magnetically constrained nanocontacts.

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Mixed valence compounds of the form $La_{1-x}A_xMnO_3$ ($A$ being Ca, Sr or Ba) have recently been shown to present extremely large (colossal) magnetoresistance. [1,2] In these materials strong Hund’s interaction between the charge carriers and the manganese ions leads to a strong coupling between the electrical resistivity and the magnetic state. For $0.1 \leq x \leq 0.5$ and low temperatures, the system is metallic and presents ferromagnetic order. As the temperature increases the system becomes insulator and paramagnetic. The magnetic transition occurs at a $x$-dependent critical temperature $T_c \sim 300K$. Colossal magnetoresistance occurs near $T_c$ and in presence of magnetic fields of several Teslas.

In the $La_{1-x}A_xMnO_3$ compounds, the electronically active orbitals are the $Mn$ $d$ orbitals, and the mean $d$ occupancy is $4 - x$. A strong ferromagnetic Hund’s rule coupling aligns all electron spins in the $Mn$ $d$ orbitals. The $Mn$ ions form a simple cubic lattice of lattice parameter $a$. The cubic crystal symmetry splits the $d$ orbitals into a $t_{2g}$ triplet and an $e_g$ doublet. Three electrons fill up the $t_{2g}$ levels forming a core spin of magnitude $S = 3/2$ and the rest of the electrons, $1 - x$ per $Mn$, go to the $e_g$ orbitals.

Ferromagnetism in these materials is explained by the Double Exchange (DE) mechanism, in which the electrons get mobility between the $Mn$ ions using the oxygen as an intermediate. [3,4] This conduction process is proportional to the electron transfer integral and due to the strong ferromagnetic Hund’s rule coupling it is maximum when the two cores spins involved in the process are parallel and it is zero when they are antiparallel. Because the alignment of spins favors electronic motion, the ferromagnetic ground state maximizes the electron kinetic energy. When the temperature increases, the DE model undergoes a phase transition towards a paramagnetic state. In this phase the core spins are randomly oriented and the electron kinetic energy is minimized. In the paramagnetic phase these materials behave as electrical insulators. [5]

Large low-field magnetoresistance has been observed in ferromagnetic $La_{1-x}A_xMnO_3$ compounds with different structural discontinuities. [6] These effects are associated with a lack of oxygen at the interface which produces an antiferromagnetic ordering at the interface and breaks the DE mechanism. [6]

Below $T_C$ these materials may contain magnetic domains separated by domain walls (DW’s). Domain walls produce a resistance to the electrical current, and for understanding low-field magnetoresistive effects in DE materials it is important to know the width and the resistance of DW’s. Domain wall magnetoresistance effects have been recently observed in itinerant ferromagnet systems such as Co, Ni and Fe [12,13] and also in colossal magnetoresistance perovskites. [13] In reference [13] it is obtained that in $La_{1-x}Ca_xMnO_3$ the resistance of a domain wall is $8 \times 10^{-14} \Omega m^2$, a quantity that the authors argue is 4 orders of magnitude larger that one might expect based in DE models. Recent theoretical works have studied the ballistic and diffusive transport through domain walls in itinerant ferromagnets, [14] however for DE systems only the transmission in one-dimensional models have been studied. [15]

In this paper we study magnetic domain walls in DE systems. In the DW the direction of the $Mn$ spin changes from 0 to $\pi$ over a region of width $L_W$. In this region the $Mn$ core spins are misaligned and the tunneling amplitude between $Mn$ ions along the DW is reduced. This loss of kinetic energy in the DE model is the equivalent to the loss of exchange energy in the Heisenberg model. The chemical potential in the system is fixed by the magnetic domains which have all the same hole concentration $x$. The reduction of the bandwidth in the DW region with respect of the surround magnetic domains produces a change in the density of electrons in the DW region. This effect cost a lot of Hartree energy and the system prefers to create dipoles at the edge of the DW’s. In this way the $Mn$ ion levels change and the local charge is not modified in the DW. The shift of the energy level of the $Mn$ ions modifies the cost of creating a DW and therefore its width. This effect is new and occurs due to the DE mechanism. In the first part of the paper we characterized this effect and study how it affects the width of the DW. In the second part of the paper we study the
ballistic transport through DW’s in DE systems.

**Microscopic Hamiltonian.** We are interested in a hole concentration in the range \(0.1 \leq x \leq 0.4\). For this doping range the \(e_g\) orbitals are degenerated and for simplicity we consider that the Hamiltonian is degenerated in the \(e_g\) orbital index. Also in our model the \(Mn\) spins are treated as classical. For temperatures below \(T_c\) and in the limit of infinite Hund’s coupling, the electronic properties of the \(Mn\) oxides are described by the nearest neighbor tight binding Hamiltonian,

\[
\hat{H}_{DE} = -\sum_{i,j,\alpha} t_{i,j} \hat{C}_{i,\alpha}^+ \hat{C}_{j,\alpha},
\]

where \(\hat{C}_{i,\alpha}^+\) creates an electron at site \(i\), in the orbital \(\alpha\) and with spin parallel to the core spin at site \(i\), and the hopping amplitude is given by

\[
t_{i,j} = t \left( \cos \frac{\theta_i}{2} \cos \frac{\theta_j}{2} + \sin \frac{\theta_i}{2} \sin \frac{\theta_j}{2} e^{i(\phi_i - \phi_j)} \right),
\]

where \(\theta_i\) and \(\phi_i\) are the angles which characterize the orientation of the core spin at site \(i\).

**Long-wavelength functional.** In a perfect system all core spins are parallel and the electron kinetic energy gets its maximum value. Now consider a modulation of the core spin direction characterized by a vector \(\mathbf{M}_j = S_j / S\). In the long-wavelength limit the modulation can be described by a continuum unitary vector field \(\mathbf{M}(\mathbf{r})\). This modulation produces a decrease of the value of the hopping amplitude, and therefore a kinetic energy loss. For smooth modulations the local loss of kinetic energy is proportional to \((\nabla \mathbf{M})^2\) and the difference in kinetic energy (KE) between the uniform system and the modulated system is given by

\[
\Delta E^{KE} = \frac{\rho^{KE}}{2} \int d^3 \mathbf{r} (\nabla \mathbf{M})^2.
\]

Here \(\rho^{KE}\) is the KE stiffness of the system. This term is the equivalent to the term describing the exchange energy loss in the Heisenberg model. Note, however that in the DE system \(\rho^{KE}\) is associated with the loss of electron kinetic energy due to the spatial variations of \(\mathbf{M}\). By diagonalizing the Hamiltonian Eq.1 with different boundary conditions we have calculated the value of \(\rho^{KE}\) for different hole concentration in the system. In Fig.1 we plot \(\rho^{KE}\) as a function of \(x\). Note that we have a degeneration 2 associated with the \(e_g\) orbitals and we measure the hole concentration with respect half-filling. When the electron concentration is zero \((x = 1)\) the band is completely empty there is not KE in the system and \(\rho^{KE} = 0\). When the band is half filled \((x = 0)\) the KE is maximum and \(\rho^{KE}\) gets its maximum value.

Besides the KE, there is also a Hartree (H) term which contributes to the long-wavelength functional. The electron density, \(n\) , in different regions of the sample is fixed by the chemical potential \(\mu\) which is obtained from the value of \(n\) in the regions of constant magnetization (magnetic domains). The tunneling amplitude only depends on the relative orientation of the \(Mn\) spins and therefore \(n\) and \(\mu\) get the same values within all the magnetic domains. These regions represent basically all the system and they can be considered as reservoirs for the electrons. The total charge in the system should be zero and a background of positive charge equal to \(1 - x\) is assumed to exist in the sample.

In regions with modulated magnetization, \(\nabla \mathbf{M} \neq 0\), the tunneling amplitude is reduced and the bandwidth is narrowed. Since \(\mu\) is fixed by the reservoirs, the decrease of the bandwidth would produce a change in \(n\) in these regions. This breaks local charge neutrality and it would cost a lot of Hartree energy. The system prefers to create dipoles in order to shift the \(Mn\) ion energy levels in such a way that local charge neutrality is recovered. The energy shift is negative (positive) for electron concentration smaller (bigger) than half-filling. For smooth modulation of the magnetization, we have calculated from the Hamiltonian Eq.(1) the energy shift required for keeping local charge neutrality. We find that the shift is proportional to \((\nabla \mathbf{M})^2\) and therefore the local charge neutrality constrain gives a contribution to the energy of the system of the form,

\[
\Delta E^H = \beta \int d^3 \mathbf{r} (\nabla \mathbf{M})^2.
\]

In Fig.1 we plot the value of \(\beta\) as a function of \(x\), as obtained by solving the Hamiltonian Eq.(1) with different boundary conditions. In principle we should add also a term describing the energy cost of creating the dipoles responsible for the energy shifts, however we find that this energy is much smaller than \(\Delta E^H\).

Finally we need to know the energy term corresponding to the constrain which creates the magnetic domains. In general this term has the form,

\[
\Delta E^C = \int d^3 \mathbf{r} f(\theta).
\]

For uniaxial crystals \(f(\theta) = K \sin^2(\theta)\) being \(K\) the anisotropy constant. For domain walls created by magnetic constrictions \([12,13]\) \(f(\theta) = g \mu_B H S / a^3 \cos \theta \text{sgn}(x)\), being \(H\) the magnetic field.

Adding the different contributions we get the functional

\[
F = \frac{\rho}{2} \int d^3 \mathbf{r} (\nabla \mathbf{M})^2 + \int d^3 \mathbf{r} f(\theta).
\]

being \(\rho = \rho^{KE} + 2\beta\).

**Domain wall width.** A DW along the \(\hat{x}\) direction has the general form \(\mathbf{M} = (0, \sin \theta, \cos \theta)\) and \(\cos \theta(x)\) is obtained by minimizing \(F\) with the boundary conditions,
\( M \to \pm \hat{z} \) at \( x \to \pm \infty \). For uniaxial crystals the optimum form of the DW is

\[
\cos \theta = -\tan h \frac{4x}{L_W} \tag{7}
\]

with \( L_W = \sqrt{\mu/2K} \). For a magnetic constrain the form of the domain wall is given by

\[
\cos \theta = \left\{ 1 - 2 \cosh^{-2} \left[ \frac{4x}{L_W} + \ln(\sqrt{2}+1) \right] \right\} \text{sgn}(x) \tag{8}
\]

and \( L_W = 4\sqrt{\frac{a^2}{\mu g_H H_S}} \). In both cases \( L_W \) represents the width of the DW, and it is proportional to the square root of the total stiffness \( \mu \). The effect of the Hartree term to the DW width depends on the electron concentration: for electron concentrations smaller than half filling \( \beta \) is negative and the Hartree term prefers thin DW’s. On the contrary for electron concentrations bigger than half filling \( \beta \) is positive and the Hartree term likes wide DW’s.

For a given value of \( L_W \) equations (7) and (8) have essentially the same form and for simplicity we use expression (7) for describing a domain wall.

**Transport through a domain wall.** Now we calculate the ballistic conductance, \( G \), associated with a DW. From the difference between the conductances of a perfect system and a system with a DW we can evaluate the low field magnetoresistance associated with the alignment of the magnetic domains. A DW modulates the magnetization only along \( \hat{x} \) direction and the Hamiltonian is invariant in the \( \hat{y} - \hat{z} \) plane. Therefore the conductance of the system can be written as

\[
G(\mu) = \int dE' g_{1D}(E') n^{2D}(\mu - E') , \tag{9}
\]

here \( n^{2D}(E) \) is the two-dimensional density of states per unit area at energy \( E \), and \( g_{1D}(E) \) is the conductance of a one dimensional system at energy \( E \). Expression (9) can be interpreted as the sum of the conductance of all the one dimensional channels. The one dimensional conductance between the sites 1 and \( N \) is written as,

\[
g_{1D}(\mu) = \frac{2e^2}{\hbar} 4\pi^2 t^4 D(\mu)|G_{N,1}(\mu)|^2 \tag{10}
\]

being \( D(\mu) = \sqrt{4t^2 - \mu/(2\pi t^2)} \) the local density of states at an edge of the isolated lead, and \( G_{N,1}(\mu) \) the Green function connecting the sites 1 and \( N \) of an infinite one dimensional system. The factor 2 in Eq.(10) corresponds to the \( e_g \) degeneracy. The DW is fully contained between the sites 1 and \( N \).

In order to calculate the effect of the DW on the transport properties it is necessary to know the effect that the magnetization, \( M = (0, \sin \theta, \cos \theta) \) has on the electron Hamiltonian. There are two effects: the modulation of the hopping amplitude along the \( \hat{x} \) direction and the shift of the \( Mn \) ion levels. Both effects are obtained from the form of the DW, Eq.(7).

From Eq.(9) we evaluate the magnetoresistance,

\[
MR = \frac{G_0 - G_{DW}}{G_0} \tag{11}
\]

Here \( G_{DW} \) and \( G_0 \) are respectively the conductance in presence and in absence of the DW. \( MR \) represents the low-field ballistic magnetoresistance associated with the presence of magnetic domains. In Fig.2 we plot, for different hole concentrations, \( MR \) as a function of \( L_W \). For small values of \( L_W \), \( MR \) gets rather large values (> 10%). For \( L_W \geq 20a \), the magnetoresistance is always smaller than 1%. In the inset of Fig.2 we plot the the one-dimensional conductance, \( g_{1D} \) as a function of the energy for a DW with \( L_W = 5a \). The asymmetry with respect to zero energy, is due to the Hartree energy level shift. We see that \( g_{1D} \) is suppressed mainly at the band edges.

This is the reason why the \( MR \) is bigger for small concentration of electrons, where the Fermi energy is close to the band edge.

**Discussion.** We now discuss the application of our results to manganese oxides. First we want to know the width of the DW produced by crystal anisotropy. The width is determined by the stiffness, \( \rho \), and by the anisotropy constant \( K \). \( K \) can be obtained experimentally from neutron scattering [19–21], by microwave absorption [22] and by studies of the resistance saturation with magnetic fields. From these experiments we estimate an anisotropy constant in the range \( 2 \times 10^5 - 5 \times 10^6 Jm^{-3} \), for \( x = 0.3 \) this implies a DW width in the range \( 10a \leq L_W \leq 30a \) and a ballistic MR of 1 − 2%.

In the case of DW’s created by magnetic constrictions [13], \( L_W \) depends on the applied magnetic fields. For \( x = 0.3 \), we obtain \( L_W \approx 26a/\sqrt{H} \) being \( H \) the external magnetic field in Teslas. This corresponds to a rather large DW. Therefore we expect that the DW width should be determined by the crystal anisotropy and \( \sim 1 - 2\% \) magnetoresistances are expected. This value is similar to the obtained in reference [13], assuming \( 10a \leq L_W \leq 30a \). However we do not know what is the contribution of diffusive processes to \( MR \). Within the simplest Born approximation, diffusive effects are described by the reduction of the bandwidth, and for the values of \( L_W \) considered this produces also a 1% magnetoresistance.

From the inset of figure 2, we can say that the magnetoresistive effects should be much more important in geometrically constrained domain walls. Recently magnetoresistances of 200% have been observed in nanoco-ntacts of itinerant ferromagnets. We expect similar effects to occur in DE materials confined to reduced dimensions.

**Summary.** We have calculated the width of domain walls in double exchange materials. The width is propor-
tional to the square root of the stiffness. The stiffness has two terms; the kinetic energy and the Hartree term. The kinetic energy is the equivalent to the exchange energy in the Heisenberg model and it comes from the decrease of the tunneling amplitude in the domain wall region. The Hartree term appears only in double exchange materials and it comes from the connection between band-width and magnetization. We have also calculated the low-field magnetoresistance associated with the existence of magnetic domains. We have found a magnetoresistance of 1-2%. The magnetoresistance can be considerably larger in magnetically constrained nanocontacts.

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