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Tunnelling magnetoresistance in nanometer granular perovskite systems

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Abstract. In this contribution the phenomenological theory for the tunnelling magnetoresistance phenomenon observed in granular perovskite manganese systems is developed using Landauer ballistic transport concept. It was shown that the field dependence, magnitude and derivative of magnetoresistance ratio observed experimentally are well reproduced by the presented theory.

Keywords: Tunnelling magnetoresistance, perovskite.

1. Introduction
Tunneling magnetoresistance (TMR) in various spintronic materials including manganese perovskites Ln1−xAxMnO3 (Ln and A are rare earth and alkaline ions) and structures has involved much attention of researchers during the last decade (see the review [1]). It was shown in Ref. [2] that spin polarized tunneling transport is the dominated mechanism of magnetoresistance effect in nanometer perovskite maganates. In earlier work [3], Hwang et al. suggested that magnetoresistance in polycrystalline perovskite La0.7Sr0.3 MnO3 originates from the following two sources: (i) an intrinsic part arising from the double exchange mechanism between two neighboring manganese ions and (ii) the intergranular spin-polarized tunneling at the grain boundaries. Both these factors were considered in the theory developed in Ref. [4] for spin-polarized tunneling in granular perovskites. It was shown in many experiments that spin polarized tunneling gives rise to a sharp drop in resistance at low fields and low temperature. Inoue and Maekawa [5] had applied the Landauer ballistic transport concept [6] for description of tunneling conductance $G$, where it is given by

$$G = \frac{e^2}{h} |T|^{2}$$

(1)

In this equation $e$, $h$ are the electron electric charge and Planck constant, and $|T|^{2}$ is transmission coefficient of an electron wave crossing intergranular potential barrier (see figure 1). Let $\chi$ be the angle between the magnetizations of nearest-neighbor grains (see figure 1), one can write the electron transmission coefficient in the following form (see Ref. [4])

$$|T|^{2} \approx \frac{1}{2} \left( n_{t} + n_{\perp} \right)^{2} \left[ 1 + P^{2} \cos \chi \right]$$

(2a)

$$P = \frac{n_{t} - n_{\perp}}{n_{t} + n_{\perp}}$$

(2b)
where \( P \) is the electron polarization. Since the electrostatic grain charging energy \( E_C = e^2 / 2C \) (\( C \) is the grain capacitance) and the distance \( s \) between grains, also influence on the electron tunneling the conductance between grains can be expressed as \[5\]

\[
G_{ij} \propto \left( 1 + P^2 \cos \chi \right) \exp \left(-\frac{E_C}{k_B T} - 2ks \right)
\quad (3a)
\]

\[
k = \left( 2m_e \phi / \hbar \right)^{1/2}
\quad (3b)
\]

In (3b) \( m_e, \phi \) are the effective mass of electron and the potential barrier height, respectively. Influence of the intergranular distance and charging Coulomb energy on the electron tunneling were discussed also in \[7\], \[8\].

Figure 1. The potential barrier model for the electron tunneling with the spin rotation occurred between two spherical magnetic grains.

Helman and Abeles \[8\] showed that the product of \( sE_C \) is constant for the certain systems

\[
sE_C = c
\quad (4)
\]

Here \( c \) is constant. Taking into account the relation (4) one should average the tunneling conductivity (3a) over directions of grain magnetizations and distribution of distance \( s \) between grains as follows \[5\]

\[
G_{av} (H) = \int \int \int d\Omega_1 d\Omega_2 f (s) g (\theta, H) \left( 1 + P^2 \cos \chi \right) \exp \left(-\frac{c}{sk_B T} - 2ks \right)
\quad (5a)
\]

where \( d\Omega = \sin \theta d\theta d\phi \) is the element of solid angle. \( f (s), g (\theta, H) \) are specific distribution functions for the intergranular distance and grains orientations in the external magnetic field \( H \). Its explicit forms will be given later. The function \( \cos \chi \) in (5a) – the cosine of the angle between grain magnetization \( \vec{M}_1 (M, \phi_1, \theta_1) \) and \( \vec{M}_2 (M, \phi_2, \theta_2) \) is expressed through the spherical coordinate angles as:

\[
\cos \chi = \sin \theta_1 \sin \theta_2 \cos (\phi_1 - \phi_2) + \cos \theta_1 \cos \theta_2
\quad (5b)
\]

In \[5\] the electron polarization \( P \) was considered simply as a parameter of the theory. In more realistic way, the authors \[4\] calculated \( P \) within the molecular field approximation for the localized spins inside the grains and \( P \) was equal to the electron relative magnetization \( m \).

The aim of this contribution is to develop further a phenomenological theory for the TMR in granular perovskite systems basing on Inoue-Maekawa theory \[5\]. We will show that in the case of perovskites, when there is strong local Hund interaction (\( J_H \)) between electron and localized spins (\( J_H >> t, t \) is an electron hopping integral), the electron polarization \( P \) can reach almost saturated values. After that, we will calculate conductivity (formula (5a)) with reasonable
proposed $f(s), g(\theta, H)$ functions and estimate the TMR effect for nanometric granular perovskite systems. This theory also can be applied for different perovskite manganese systems when the condition of strong coupling between tunneling electron and localized spin inside nanometer grains is satisfied.

2. Electron polarization
In order to describe the electron polarization in perovskite manganese grains one can use the Kondo lattice model as follows

$$H = \sum_{\langle ij \rangle} t_{ij} a_{i\lambda}^\dagger a_{j\lambda} - J_H \sum_{j,\alpha\beta} \vec{S}_j a_{i\alpha}^\dagger(\sigma_{\alpha\beta}) a_{i\beta} - \frac{1}{2} \sum_{\langle ij \rangle} I_{ij} \vec{S}_i \cdot \vec{S}_j$$

where $a_{i\lambda}^\dagger(a_{j\lambda})$ is the creation (annihilation) electron operator at site $i$ with spin projection $\lambda = \uparrow, \downarrow$ or $\pm 1$. $t_{ij}$ is the nearest-neighbor hopping energy of the $e_g$ electron. $J_H$ is the local ferromagnetic Hund’s rule coupling between the $e_g$ electron spin $\sigma/2$ and the $t_2g$ localized spin $S$ at $i$-th site. The last term in (6) corresponds to the antiferromagnetic exchange between nearest neighbor $t_2g$ spins with the strength $I_{ij}$. Because of the strong onsite Hund exchange interaction between the tunneling electron and the localized $t_2g$ spin $S = 3/2$, then the shift of the electron band caused by this interaction is primary important comparing with a grain size effect. For the bulk materials, it was shown by our temperature dependence Green function method calculation [9] that the electron dispersion law in the strong coupling limit ($J_H \gg H$) is given approximately by

$$E_{\epsilon_\lambda} = -\lambda J_H \left(S + \frac{1}{2}\right) + \left(1 + \frac{1}{2S}\right)t_i$$

It is obviously that when $J_H \gg t$, the second term in (7) can be neglected. The electron number $n_{\epsilon_\lambda} = \frac{1}{N} \sum_{i} n_{i\epsilon_\lambda}$ figured in the expression for the electron polarization $P$ is derived according to the common formula:

$$n_{i\epsilon_\lambda} = \langle a_{i\lambda}^\dagger a_{i\lambda} \rangle = \lim_{\gamma \to 0} \int dE \left\langle \left\langle a_{i\lambda}^\dagger a_{i\lambda} \right\rangle_{E_{\epsilon_\lambda} \gamma} \right\rangle f(E)$$

where $f(E) = \{\exp[(E - \mu)/k_BT]\}^{-1}$ is the Fermi-Dirac distribution function and $\left\langle \left\langle a_{i\lambda}^\dagger a_{i\lambda} \right\rangle_{E_{\epsilon_\lambda} \gamma} \right\rangle$ is the retarded Green function given in the energy representation. The electron polarization is derived from (7), (8) and (2b):

$$P = \tanh \left[ \frac{J_H}{k_BT} \left(S + \frac{1}{2}\right) \right]$$

It follows from (9) that at low temperature, where the electron tunneling phenomenon in perovskites is more interest, the electron polarization $P$ may have values closed to the saturated one (hundred percents).

3. Tunneling magnetoresistance
The tunneling magnetoresistance ratio (MR) according to (5a) is defined by the equation

$$MR = \frac{\rho(H) - \rho(0)}{\rho(0)} = \frac{G_{av}(0) - G_{av}(H)}{G_{av}(0)}$$

where $G_{av}(H)$ ($G_{av}(0)$) is electron tunneling conductivity in the external field $H$ (without the external field $H$) and $G_{av}(H) = \rho^{-1}(H)(G_{av}(0) = \rho^{-1}(0))$. The distribution function for the intergranular distance is proposed to be:
where \( s_0 \) is the distance between grains which causes maximum of the electron tunneling conductivity. The grain orientation at finite temperature in the actual field is defined with statistical factor \( g(\theta, H) \)

\[
g(\theta, H) = \frac{1}{A} \exp \left( \frac{M (H - N_d M) (\cos \theta_1 + \cos \theta_2)}{k_B T} \right)
\]

here \( N_d \) means the demagnetization factor for spherical shape grains. \( N_d \) equals to \( 4\pi/3 \) (0.33) in CGS (SI) system of units. For small ferromagnetic grains, especially in nanometer range, the demagnetization field effect should be very important and an actual field inside grains acting on electron may be reduced strongly comparing with the applied field \( H \). In the equation (12), the demagnetization field is written as \( H_{N_d} = -N_d M \) and \( A \) stands for a normalized constant

\[
A = 4\pi^2 \left[ \frac{\sinh \left( \frac{M (H - N_d M)}{k_B T} \right)}{M (H - N_d M)} \right]^{-1/2}
\]

Inoue and Maekawa [5] used the rough approximation when \( \cos \chi \) is replaced by the square of relative magnetization i.e. \( \cos \chi = M^2 \). In the work [4], \( \cos \chi \) was considered as an adjust parameter. Here we can calculate directly without these assumption and get the following expression for the average tunneling conductance

\[
G_a(H) = \frac{2c G_0 K_2}{s_0 k_B T (2ks_0 + 1)} \left[ 1 + P^2 \left[ \coth \left( \frac{M (H - N_d M)}{k_B T} \right) - \frac{k_B T}{M (H - N_d M)} \right]^2 \right]
\]

where \( K_2(x) \) is McDonal’s function.

The magnetoresistance ratio is obtained straightforwardly from (15) and (10):

\[
MR = -\frac{P^2}{1 + P^2} \left[ \coth \left( \frac{M (H - N_d M)}{k_B T} \right) - \frac{k_B T}{M (H - N_d M)} \right]^2
\]

Proposing that at low fields, the magnetization of the nanometric perovskite grains system has the linear dependence on the field intensity, \( M = \kappa H \), where \( \kappa \) is the magnetic susceptibility, we get
\[
MR = \frac{\tanh^2 \left( \frac{\alpha}{H^2_0} \right)}{1 + \tanh^2 \left( \frac{\alpha}{H^2_0} \right)} \left[ \coth \left( \frac{H}{H_0} \right) - \left( \frac{H_0}{H} \right)^2 \right] 
\]

(17)

where \( \alpha = J_H \left( S + \frac{1}{2} \right) \) and \( H_0 = \left( \frac{k_B T}{\kappa (1 - N_d \kappa)} \right)^{1/2} \).

Figure 2. (a) The magnetoresistance ratio MR as a function of the reduced external magnetic field; (b) The field dependence of the derivative of MR. Here \( \alpha / H^2_0 = 0.96 \)

Figure 2a shows the negative MR given by the formula (17) as a function of inverse relative field strength. We noted that a shape of this curve and a saturated value of MR were the same as observed in experiments [2, 4]. Figure 2b demonstrates the field dependence of the derivative of the MR ratio. The ABC parts of the plots given in figure 2 have the same shape compared with experimental curves obtained in many works, and the 0A parts were not analyzed experimentally. Figure 3 shows the comparison between our theory and experimental results for low field MR (figure 3a), and its derivative (figure 3b) registered in nanometric granular perovskite La_{0.67}Sr_{0.33}MnO_3 at temperature 80, 150K (see Dey et al. [10]). One can find here an excellent agreement between the theoretical and experimental results.

Figure 3. Comparison between theory and experimental results for low field MR (a) and its derivative (b) given in Ref. [10]. Here the parameter \( \alpha = 1.17; H_0 = 1.43 \) and \( \alpha = 2.00; H_0 = 1.89 \) for T = 80 K and T = 150 K, respectively. An average diameter of grains is D = 27 nm.
In conclusion, we have studied the low field tunneling spin transport in magnetic nanometer granular perovskite systems and have shown that the strong interaction between the electron spin and localized atomic spins inside grains leads to the high polarization degree of tunneling electrons. The field dependence of the TMR in nanometer perovskite, its magnitude and derivative can be described properly in the framework of our theory.

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