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A simple formulation for magnetoresistance in metal-insulator granular films with increased current

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We studied the tunnel magnetoresistance in metal/insulator granular films when the applied current is varied. The tunnel magnetoresistance shows a strong modification related to a non-Ohmic behaviour of theses materials. It was verified that spin-dependent tunnelling is the main mechanism for magnetoresistance at low applied current. However, when the current is high, another mechanism gets to be important: it is independent of the magnetization and is associated to variable range hopping between metallic grains. In this work, we propose a simple modification of Inoue and Maekawa's model for tunnelling magnetoresistance in granulars, rewriting the expression for resistance as a function of magnetic field and temperature, also taking into account the two different contributions. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4793272]

I. INTRODUCTION

The metal/insulator granular systems, i.e., composites formed of metallic grains embedded in an insulator matrix,[1,2] have been extensively studied by many groups due to the rather interesting phenomena observed, like tunnel magnetoresistance (MR),[3–5] among others.

In metal/insulator granular systems, the electrical current should be due to electrons tunnelling between grains. If the metallic grains are ferromagnetic, tunnel magnetoresistance is expected to be present.

The tunnel magnetoresistance is associated to spin-dependent tunnelling, i.e., the probability of electron tunnelling is higher when the magnetic moments of the grains are parallel rather than anti-parallel, and this effect is well explained by the model of Inoue and Maekawa.[6]

There are other models for granular ferromagnetic magnetoresistance[7] that consider the spin-dependent tunnelling and study the magnetoresistance in both the low-field ($e\Delta V \ll k_BT$) and high-field regimes ($e\Delta V \gg k_BT$).

In previous work,[8] we presented results of the modification of magnetoresistance in Fe–Al_2O_3 granular films when the applied current or bias potential are increased, which cannot be explained by spin-dependent tunnelling only or by the models above.[6,7]

Beloborodov *et al.*'s model for nanogranular ferromagnetic magnetoresistance[7] describes the non-linear resistance in the high-field regime. What we propose with our model is the study of the variation of the resistance, and even the inversion of the magnetoresistance, in the low-field regime, with changing current or bias.

Moreover, the expression for the magnetoresistance found in the literature[7] does not describe its inversion, as seen in our previously published experimental data.[8]

In this work, we aim at presenting a discussion, based on experimental results, of a simple modification to the model of Inoue and Maekawa,[6] that is able to explain the dependence of the tunnelling magnetoresistance when the current is high as well as an expression for electrical resistance as function of magnetic field and temperature (as suggested in Ref. 8), starting from very basic considerations.

II. DEVELOPMENT/ANALYSIS

For variable range hopping, the wave function of an electron tunnelling between two states in metallic grains separated by an insulating barrier is

$$\psi(r_{ij}) \propto \exp\left(-\frac{r_{ij}}{\xi}\right), \tag{1}$$

where r_{ij} is the hopping distance between the two grains, $\xi = \hbar/(2\ m|\varepsilon|)^{1/2}$ is the localization length, $\varepsilon < 0$ and m is the effective mass.[11]

Inoue and Maekawa[6] consider, when the electron is tunnelling between two ferromagnetic grains, that this tunnelling depends on the angle between the magnetic moments of the grains, that is, spin-dependent tunnelling.

The hopping resistance of an electron tunnelling between two localized states in ferromagnetic grains can thus be written as

$$R_{ij} \propto \frac{1}{(1 + P_iP_j\cos\theta)}\exp\left(2\frac{r_{ij}}{\xi} + \frac{\varepsilon_{ij}}{k_BT}\right), \tag{2}$$

where θ is the angle between the magnetic moments of the grains, k_B is Boltzmann's constant, T is the temperature, $\varepsilon_{if} = 1/2\ (|\varepsilon_i - \varepsilon_F| + |\varepsilon_j - \varepsilon_F| + |\varepsilon_i - \varepsilon_j|)$, ε_i and ε_j are the energies of states i and j, ε_F is the Fermi energy, and P is the spin polarization in the ferromagnetic grains.

The polarization is given by $P = \frac{D_\uparrow - D_\downarrow}{D_\uparrow + D_\downarrow}$, with the density of states at the Fermi energy D_σ ($\sigma = \uparrow,\downarrow$). P_i is the spin polarization of grain i and P_j is the spin polarization of grain j.

A magnetic field can greatly alter the asymptotic behaviour of the wave function[11] for an electron tunnelling between two localized states; consequently, we have

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$$\psi(r_{ij}) \propto \exp\left(-\frac{r_{ij}}{\xi} - f(H)\right), \tag{3}$$

where $f(H)$ is a function of the magnetic field that changes the wave function, and H is the magnetic field, whose direction is in the z-axis.

For $\lambda \gg \xi$ and $\rho \ll \lambda^2/\xi$, the function $f(H)$ is $\frac{r_{ij}\rho^2\xi}{24\lambda^4}$, where $\lambda = (c\hbar/e\,H)^{1/2}$ is the magnetic length, and $\rho = r_{ij}\sin\gamma$ is the distance from $\vec{r_{ij}}$ to the z-axis.[11]

Considering the spin-dependent tunnelling and the change of behaviour of the wave function due to the magnetic field, the electrical resistance between two ferromagnetic grains will be given by

$$R_{ij} \propto \frac{1}{(1 + P_i P_j \cos\theta)} \exp\left(2\frac{r_{ij}}{\xi} + \frac{\varepsilon_{ij}}{k_B T} + 2f(H)\right). \tag{4}$$

The angle θ and the hopping distance r_{ij} are randomly distributed in the metal/insulator granular films, so one can take an average over these quantities.

Also, the resistance of a ferromagnetic metal/insulator granular system should take into account all possible distances between grains and all angles between the magnetic moments of grains. So we write

$$R = R_0 \int\int f(r) f(\theta) \frac{1}{(1 + P_i P_j \cos\theta)} \times \exp\left(2\frac{r}{\xi} + \frac{\varepsilon_{ij}}{k_B T} + 2f(H)\right) d\theta . dr, \tag{5}$$

where R_0 is the resistance for $H = 0$, $r = r_{ij}$, $f(r)$ is the distribution function for r, and $f(\theta)$ is the distribution function for θ.

Integrating Eq. (5) over θ, we have that the exponential is independent of the angle θ, so the resistance becomes

$$R = R_0 \int f(r) \frac{1}{(1 + P_i P_j \langle\cos\theta\rangle)} \exp\left(2\frac{r}{\xi} + \frac{\varepsilon_{ij}}{k_B T} + 2\frac{r\rho^2\xi}{24\lambda^4}\right) dr, \tag{6}$$

where $\langle\cos\theta\rangle$ is the average over θ.

Using the same supposition as Inoue and Maekawa,[6] we can write $\langle\cos\theta\rangle = m^2$, where m is the relative magnetization of the system. We have here neglected the correlation between magnetic moments of neighbouring grains.

In a superparamagnetic granulars, the relative magnetization of the system is given by a Langevin function,[12] i.e., $m = \coth(\mu H/k_B T) - k_B T/\mu H$, where μ is the magnetic moment of the grains.

In variable range hopping, it is necessary to minimize the exponent of the electrical resistance[11] (Eq. (6)). We consider that $r_{ij} \approx \xi\ (T_0/T)^{1/4}$ and $\Delta_M \approx a k_B {T_0}^{1/4} T^{3/4}$, where $\varepsilon_{ij} = \Delta_M = \frac{\Delta}{N r_{ij}^3}$, $T_0 = \beta\Delta/N\xi^3$, with N being the number of localized states randomly distributed in a band Δ centered at the Fermi level ε_F; a and β are numerical factors.

Using those parameters, we find for the exponential in Eq. (6)

$$\frac{2r_{ij}}{\xi} + \frac{\varepsilon_{ij}}{k_B T} + 2\left(\frac{r_{ij}\rho^2\xi}{24\lambda^4}\right) = \left(2 + \frac{ak_B}{k_B}\right) . \left(\frac{T_0}{T}\right)^{1/4} + \frac{\xi^4 \sin^2\gamma}{12\lambda^4} . \left(\frac{T_0}{T}\right)^{3/4} = \alpha\left(\frac{T_0}{T}\right)^{1/4} + AH^2$$

where $\alpha = (2 + a)$ and

$$A = \left(\frac{\xi^2 e \sin\gamma}{\sqrt{12} c\hbar}\right)^2 (T_0/T)^{3/4} = t\left(\frac{\xi^2 e}{c\hbar}\right)^2 (T_0/T)^{3/4}, \tag{7}$$

with $t = \frac{\sin^2\gamma}{12}$.

The expression for the resistance can now be written as

$$R = \frac{R_0 \exp(T_0/T)^{1/4}}{(1 + \langle P_i P_j \rangle m^2)} \exp(AH^2), \tag{8}$$

where $\langle P_i P_j \rangle$ is the average of the product of the spin polarizations of grains i and j. In doing so we are, in effect, considering all possible electron paths.

The constant A, as defined in Eq. (7), is positive and does not modify the value of R for zero magnetic fields. However, when H is not zero, R can increase if ξ is high enough. Also, A increases when ξ increases, or T decreases.

The equation for resistance can now be rewritten as follows:

$$R = \frac{R(0, T)}{(1 + \langle P_i P_j \rangle m^2)} \exp(AH^2), \tag{9}$$

where $R(0,T) = R_0 \exp(T_0/T)^{1/4}$, which corresponds to Mott variable range hopping.

The proposed formulation takes into account the modification of the wave function when the magnetic field is applied along with is spin-dependent tunnelling.

Equation (9) describes the two contributions to the magnetoresistance, and the influence of the applied current: when the current increases, ξ is increased and the exponential term becomes stronger. For low current, ξ is small, the tunnelling paths are small, A is approximately zero, $\langle P_i P_j \rangle \approx P^2$ and Eq. (9) will be equivalent to Inoue and Maekawa's model, however, in the variable range hopping regime.

Remark that the localization length is associated to all possible electronic paths in the sample. Then, when the current increases and new parallel paths are created, as proposed on Ref. 9, the localization length will be modified, changing the resistance accordingly.

For nonmagnetic samples, $m = 0$ and Eq. (9) will be

$$R = R(0, T) \exp(AH^2). \tag{10}$$

For $H = 0$, Eq. (10) is the variable range hopping regime equation.

As can be seen in the field dependence of Eq. (10), we obtain magnetoresistance even for non-magnetic granulars. This, to our knowledge, has not been tested experimentally to date.

III. APPLICATION To Fe-Al_2O_3

The experimental results in Fe-Al_2O_3 granular thin films[9] have shown that the transport properties are (i) better characterized by what can be recognised as Mott variable range hopping than by a thermally activated one and (ii) that when the applied current or bias potential are increased, the electrical resistance of the samples is decreased (and the localization length increased).

To illustrate these results, Fig. 1 presents lnR vs. $T^{-1/2}$ and lnR vs. $T^{-1/4}$ plots as well as a fit using thermally activated hopping for Fe-Al_2O_3 with a volume fraction of 0.41.[9] It can be seen that a $T^{-1/4}$ (Mott variable range hopping) fit adjusts data much better than $T^{-1/2}$ (either thermally activated hopping[1] or Efros–Shklowskii VRH[11]). This is better seen through Fig. 2, where a logarithmic plot[10] is presented to show that the exponent 1/4 describes better the problem of Ref. 9 for most of the temperature range considered.

Fig. 3 presents the R vs. V curve for the aforementioned Fe-Al_2O_3 ($x = 0.41$), at 273 K. The electrical resistance of the sample is reduced (and the localization length increased) when the bias potential is increased, as observed previously.[9]

To explain these results, some of us proposed that, when the current or bias is increased, new parallel electronic paths between more distant grains are activated, reducing the total resistance of the samples.[9]

It is essential to keep in mind that all the measurements referred were made in the low-field regime.[8,9] The process is identified as sequential tunnelling, and any modification of the magnetoresistance should be related to a change of the electrical properties in metal/insulator granular system, leading to say that these new paths do influence the magnetoresistance when the current is increased.

The mean level energy spacing (δ) was estimated to be around 10^{-23} J, and the charge energy (E_C) was calculated for the sample used in that paper to be around 0.575×10^{-20} J, so $E_C/\delta \sim 0.575 \times 10^3 = 575 \gg 1$. Assuming the same approximations as those in Ref. 13, this means that the charge energy is much larger than the spacing between states within the grains.

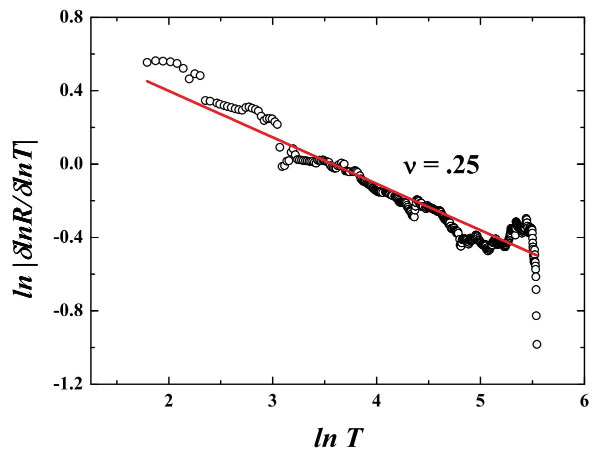

FIG. 2. The logarithmic derivative of the resistance of the Fe-Al_2O_3 granular with $x = 0.41$. The slope of the fit line is $-\nu$.[10]

Using the measurements on Fe-Al_2O_3 granular thin film[8] as an example, one observes that the magnetoresistance changes (even the sign!) when the current is increased, as shown in Fig. 4.

At $T = 200$ K, $MR = +4.4\%$ for $I = 1\,\mu$A, and $MR = -9\%$ for $I = 20$ mA. These modifications of the MR can be associated to an increase of the localization length when the current and/or the bias potential increase.

In Fig. 4, the solid lines are the fitted curves using Eq. (9) and an average magnetic moment of the grains $\mu = 2537\mu_B$ are in good agreement with the experimental results.

For low current the localization length is small and the exponential in Eq. (9) is also small. For higher currents, new paths between more distant grains are activated, so the total resistance is decreased, the localization length gets larger and the exponential is increased.

The density of states of the metallic grains should have a strong influence on the spin polarization at the Fermi

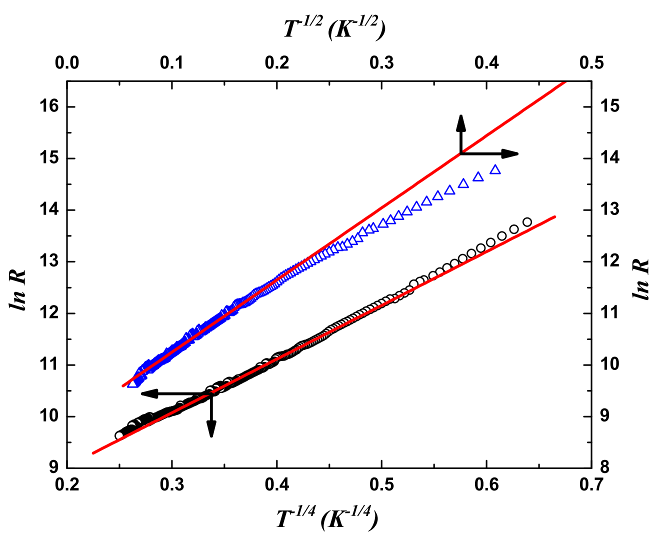

FIG. 1. (a) lnR vs. $T^{-1/2}$ plot and (b) lnR vs. $T^{-1/4}$ plots for a Fe-Al_2O_3 sample with $x = 0.41$. The solid lines are fits using $R = R_0 \exp(T_0/T)^{\nu}$ with $\nu = 1/2$ and $\nu = 1/4$.

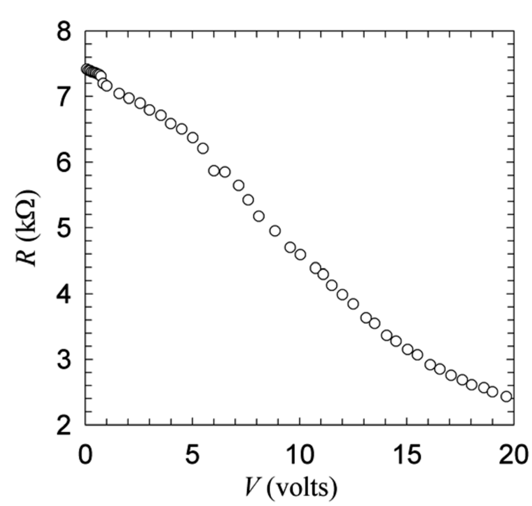

FIG. 3. R as a function of V for the sample with $x = 0.41$ at $T = 273$ K.

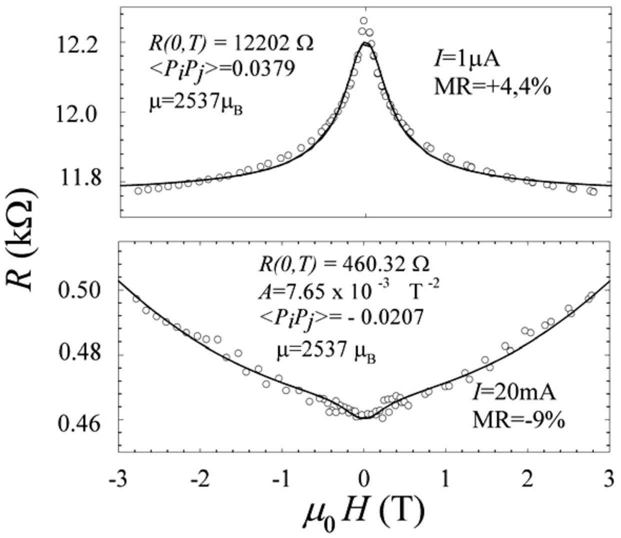

FIG. 4. R as a function of H for a Fe-Al_2O_3 granular thin film with $x = 0.41$ for different applied direct currents, at $T = 200$ K. The magnetoresistance is expressed by $MR = 100 \times (R(0) - R(H))/R(0)$. The solid lines are fits using Eq. (9) with the adjusting parameters $R(0,T)$, $\langle P_i P_j \rangle$, A, and μ.

energy. As seen Fig. 4, the proposed model explains the striking change of signal of the magnetoresistance in Fe-Al_2O_3 granular thin films.

For Fe grains in Al_2O_3, $\langle P_i P_j \rangle$ can be negative, as seen at I = 20 mA, or positive, as verified for I = 1 μA (Fig. 4). These results are carefully described in Ref. 8, and explained by the change of the Fermi level of grain j in relation to that for grain i.

The polarizations P_i and P_j are positive for hopping between nearby grains; however, P_i is positive and P_j negative for hopping between more distant grains, when the density of states for (bulk) iron is considered. So $\langle P_i P_j \rangle$ can become negative when one takes into account these new paths for high current; it all depends on the details of the density of states in case.

IV. CONCLUSIONS

The magnetoresistance in ferromagnetic metal/insulator granular thin films shows two different contributions to magnetoresistance. The first is due to spin-dependent tunnelling and the second is due to changes in variable range hopping.

For low current, only spin-dependent tunnelling is observed. When establishing higher currents, the variable range hopping contribution to the magnetoresistance is larger.

We conclude that, by taking into account both the spin-dependent tunnelling and the change imposed on the electron wave function when the magnetic field is applied, it is possible to explain the changes of the tunnel magnetoresistance when the current is varied, and propose an expression for electrical resistance as a function of magnetic field and temperature for metal/insulator granular thin films.

[1]B. Abeles, *in Applied Solid State Science: Advances in Materials and Device Research*, edited by R. Wolfe (Academic, New York, 1976) p. 1.
[2]K. M. Unruh and C. L. Chien, *in Nanomaterials: Synthesis, Properties and Applications*, edited by A. S. Edelstein and R. C. Cammarata (Institute of Physics, Bristol, 1996) pp. 129–160.
[3]T. Zhu and Y. J. Wang, Phys. Rev. B **60**, 11918 (1999).
[4]S. H. Ge, S. B. Zhang, J. H. Chi, Z. G. Zhang, C. X. Li, and R. J. Gan, J. Phys. D **33**, 917 (2000).
[5]H. Fujimori, S. Mitani, and S. Ohnuma, J. Magn. Magn. Mater. **156**, 311 (1996).
[6]J. Inoue and S. Maekawa, Phys. Rev. B **53**, R11927 (1996).
[7]I. S. Beloborodov, A. Glatz, and V. M. Vinokur, Phys. Rev. Lett. **99**, 066602 (2007).
[8]M. A. S. Boff, J. Geshev, J. E. Schmidt, W. H. Flores, A. B. Antunes, M. A. C. Gusmão, and S. R. Teixeira, J. Appl. Phys. **91**, 9909 (2002).
[9]M. A. S. Boff, F. Casarin, L. G. Pereira, and A. B. Antunes, Phys. B **406**, 1833 (2011).
[10]B. Raquet, M. N. Baibich, J. M. Broto, H. Rakoto, S. Lambert, and A. Maignan, Phys. Rev. B **65**, 104442 (2002).
[11]B. I. Shklovskii and B. Z. Spivak, in *Modern Problems in Condensed Matter Science*, edited by M. Pollak and B. Shklovskii (Elsevier Science, Amsterdam, 1991), Vol. 28, pp. 273–347.
[12]B. D. Cullity, in *Introduction to Magnetic Materials* (Addison–Wesley, USA, 1972), pp. 93–95.
[13]I. S. Beloborodov, A. V. Lopatin, V. M. Vinokur, and K. B. Efetov, Rev. Mod. Phys. **79**, 469 (2007).