Theory of giant magnetoresistance in granular alloys

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A theory of giant magnetoresistance in granular alloys was developed by considering the spin-dependent scattering within the magnetic granules and at their boundaries and the continuous distributions of granule sizes and directions of magnetization. The CPP formalism was recovered in the limit when the effective conductivities in the granules are much larger than the matrix conductivity. When this condition is not satisfied, the transport properties of the granular alloys derive from the crossover of the parallel and perpendicular current configurations. [S0163-1829(99)10309-6]

Since the discovery of giant magnetoresistance (GMR) in Fe/Cr multilayers[1] and subsequently in Co-Cu granular films,[2,3] the phenomenon has been intensively subjected to experimental and theoretical research. The mechanisms giving rise to GMR in granular alloys are the same as in layered structures, namely, the spin-dependent scattering within the magnetic granules and at their boundaries.[4] However, in granular alloys there is not such a clear distinction as the contrast in multilayers between the currents parallel (CIP) and perpendicular (CPP) to the plane of the layers. For CIP, there occurs a large magnetoresistive effect only if the mean-free path exceeds, or at least is comparable to, the thickness of the layers. This length scale does not appear for CPP, and GMR occurs even if the mean-free path is smaller than the thickness of the layers.

Zhang and Levy[4] developed a model in which the same formalism for layered structures in the CPP configuration is applied for granular alloys. The transport properties of granular alloys are modeled by considering the two spin-dependent scattering mechanisms and the distribution of sizes of the magnetic granules. In a previous paper, we examined the model in further detail.[5] Nevertheless, there is an accumulation of evidence that the size effects which are important for the CIP geometry in multilayers also contribute to the GMR in granular alloys.[6–10] When the mean-free path is comparable with the sizes and distances between the granules, current lines tend to bypass the high-resistivity granules and therefore the GMR is intermediate between CIP and CPP.

The crossover of CIP and CPP in granular alloys is accounted in classical,[6] semiclassical,[7] and quantum[8] transport theories for explaining the maximum in the annealing temperature dependence of magnetoresistance. However, the common simplifying assumptions of these treatments, namely, (i) all granules are of the same size and (ii) they have only two magnetization directions (up and down), prevent their results from comparing with the results of Zhang and Levy.

We developed a transport theory of granular alloys which, as the theory of Zhang and Levy, includes the two spin-dependent scattering mechanisms, the continuous distributions of granule sizes and magnetization directions, and as the cited theories,[6–8] takes into account the CIP and CPP mixed configurations. When the granule conductivities are much larger than the matrix conductivity, exactly the same GMR equations of Ref. 4 are obtained. In the other case when the matrix conductivity is much larger than the granule conductivities, the transport is similar to that in the CIP geometry.

So let us consider an assembly of ferromagnetic granules embedded in a nonmagnetic metal. Generally, the granules are not of the same size and the magnetic moment per granule is not constant. The density of granules whose magnetic moment is between μ and $\mu+d\mu$ is $f(\mu)d\mu$.[5] In the absence of the magnetic field $\boldsymbol{H}$, the directions of magnetization are randomly distributed.

Neglecting the spin-flip scattering, we assume that there are two independent conduction channels for spin-up (↑) and spin-down (↓) electrons. Restricting our approach to the classical electrodynamics of continuous media, we accept the local form of the Ohm's law when the quantization axis is parallel to the magnetic moment of the granule. When the absolute quantization axis is parallel to the applied magnetic field, the electric fields and the current densities take the form (Appendix)

$$\boldsymbol{E}_{\uparrow\downarrow}(\boldsymbol{r})=\frac{1}{2}\boldsymbol{E}_{+-}(\boldsymbol{r})(1+\cos\theta)+\frac{1}{2}\boldsymbol{E}_{-+}(\boldsymbol{r})(1-\cos\theta), \quad (1)$$

$$\boldsymbol{j}_{\uparrow\downarrow}(\boldsymbol{r})=\frac{1}{2}\sigma_{+-}\boldsymbol{E}_{+-}(\boldsymbol{r})(1+\cos\theta)+\frac{1}{2}\sigma_{-+}\boldsymbol{E}_{-+}(\boldsymbol{r})(1-\cos\theta), \quad (2)$$

in which θ is the angle between the magnetic moment of the granule and the magnetic field, $\boldsymbol{E}_+$ and $\boldsymbol{E}_-$ are the electric fields and σ_+ and σ_- are the conductivities of the spin-up and spin-down electrons, respectively, when the magnetic moment of the granule is parallel to the magnetic field.

Following closely the steps of Refs. 6 and 12, we express the volume average of the electric fields and the current densities in the form

$$\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_V = \frac{1}{I_s}\int_0^\infty (\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_\mu} + \langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{T_\mu})\mu f(\mu)d\mu + (1-f)\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_m}, \tag{3}$$

$$\langle \boldsymbol{j}_{\uparrow\downarrow}\rangle_V = \frac{1}{I_s}\int_0^\infty \langle \boldsymbol{j}_{\uparrow\downarrow}\rangle_{V_\mu}\mu f(\mu)d\mu + (1-f)\sigma_m\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_\mu}, \tag{4}$$

in which I_s and V_μ are the magnetization and the volume of the granule $(\mu = I_s V_\mu)$, T_μ is the volume of the transition interface between the granule and the matrix, V_m and σ_m are the volume and the conductivity of the matrix, and $1-f = V_m/V$.

The granule and matrix volume averages are defined by

$$\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_\mu} = \frac{1}{V_\mu}\int_{V_\mu} \boldsymbol{E}_{\uparrow\downarrow}(\boldsymbol{r})d^3r, \tag{5}$$

$$\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{T_\mu} = \frac{1}{V_\mu}\int_{T_\mu} \boldsymbol{E}_{\uparrow\downarrow}(\boldsymbol{r})d^3r, \tag{6}$$

$$\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_m} = \frac{1}{V_m}\int_{V_\mu} \boldsymbol{E}_{\uparrow\downarrow}(\boldsymbol{r})d^3r, \tag{7}$$

$$\langle \boldsymbol{j}_{\uparrow\downarrow}\rangle_{V_\mu} = \frac{1}{V_\mu}\int_{V_\mu} \boldsymbol{j}_{\uparrow\downarrow}(\boldsymbol{r})d^3r. \tag{8}$$

The average conductivity is the sum of the conductivities for the spin-up and spin-down channels,

$$\sigma = \tilde{\sigma}_\uparrow + \tilde{\sigma}_\downarrow, \quad \tilde{\sigma}_{\uparrow\downarrow} = \frac{\langle \boldsymbol{j}_{\uparrow\downarrow}\rangle_V}{\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_V}. \tag{9}$$

The thermal average is obtained by substituting $m(\mu,H,T)$ for $\cos\theta$ in the above equations. If the granules are superparamagnetic, the component of magnetization along the magnetic field is given by the Langevin function,

$$m(\mu,H,T) = \coth\left(\frac{\mu H}{kT}\right) - \frac{kT}{\mu H}. \tag{10}$$

Solving the classical problem of a magnetic sphere embedded in a nonmagnetic metallic matrix, we can obtain the granule averages in the Lorentz field approximation. The solution is[13]

$$\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_\mu} = \left\{\frac{1}{2}A_{+-}(\mu)[1+m(\mu,H,T)] + \frac{1}{2}A_{-+}(\mu)[1-m(\mu,H,T)]\right\}\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_m}, \tag{11}$$

$$\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_\mu} + \langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{T_\mu} = \left\{\frac{1}{2}[1+D_{+-}(\mu)][1+m(\mu,H,T)] + \frac{1}{2}[1+D_{-+}(\mu)][1-m(\mu,H,T)]\right\} \times\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_m}, \tag{12}$$

$$\langle \boldsymbol{j}_{\uparrow\downarrow}\rangle_{V_\mu} = \left\{\frac{1}{2}\sigma_{+-}A_{+-}(\mu)[1+m(\mu,H,T)] + \frac{1}{2}\sigma_{-+}A_{-+}(\mu)[1-m(\mu,H,T)]\right\}\langle \boldsymbol{E}_{\uparrow\downarrow}\rangle_{V_m}, \tag{13}$$

in which

$$\sigma_{+-}A_{+-}(\mu) = \frac{3\sigma_m}{1+2k_{+-}^{\rm CPP}(\mu)},$$

$$1+D_{+-}(\mu) = \frac{3k_{+-}^{\rm CPP}(\mu)}{1+2k_{+-}^{\rm CPP}(\mu)}, \tag{14}$$

with

$$k_{+-}^{\rm CPP}(\mu) = \frac{\sigma_m}{\sigma_{+-}}(1+r_{+-}\sigma_{+-}\mu^{-1/3}). \tag{15}$$

The coefficients r_{+-} are the effective resistivities of the interface between the granules and the matrix. Since the conductivity for the minority-spin electrons is smaller than the conductivity for the majority-spin electons, $k_-^{\rm CPP}(\mu) > k_+^{\rm CPP}(\mu)$.

Therefore, the conductivities take the form

$$\tilde{\sigma}_{\uparrow\downarrow} = \frac{\frac{1}{I_s}\int_0^\infty\left\{\frac{1}{2}\,\frac{3}{1+2k_{+-}^{\rm CPP}(\mu)}[1+m(\mu,H,T)] + \frac{1}{2}\,\frac{3}{1+2k_{-+}^{\rm CPP}(\mu)}[1-m(\mu,H,T)]\right\}\mu f(\mu)d\mu + 1 - f}{\frac{1}{I_s}\int_0^\infty\left\{\frac{1}{2}\,\frac{3k_{+-}^{\rm CPP}(\mu)}{1+2k_{+-}^{\rm CPP}(\mu)}[1+m(\mu,H,T)] + \frac{1}{2}\,\frac{3k_{-+}^{\rm CPP}(\mu)}{1+2k_{-+}^{\rm CPP}(\mu)}[1-m(\mu,H,T)]\right\}\mu f(\mu)d\mu + 1 - f}\,\sigma_m \tag{16}$$

and are proportional to the matrix conductivity.

Introducing the positive terms

$$\eta_0=\frac{2\sigma_m}{c_0}\left\{\frac{1}{I_s}\int_0^\infty\frac{3}{2}\left[\frac{1}{1+2k_-^{\mathrm{CPP}}(\mu)}+\frac{1}{1+2k_+^{\mathrm{CPP}}(\mu)}\right]\times\mu f(\mu)d\mu+1-f\right\},\quad (17)$$

$$\xi_0=\frac{1}{\eta_0}\left\{\frac{1}{I_s}\int_0^\infty\frac{3}{2}\left[\frac{k_-^{\mathrm{CPP}}(\mu)}{1+2k_-^{\mathrm{CPP}}(\mu)}+\frac{k_+^{\mathrm{CPP}}(\mu)}{1+2k_+^{\mathrm{CPP}}(\mu)}\right]\times\mu f(\mu)d\mu+1-f\right\},\quad (18)$$

$$\xi_1=\frac{3}{2\eta_0}\frac{1}{I_s}\int_0^\infty\left[\frac{k_-^{\mathrm{CPP}}(\mu)}{1+2k_-^{\mathrm{CPP}}(\mu)}-\frac{k_+^{\mathrm{CPP}}(\mu)}{1+2k_+^{\mathrm{CPP}}(\mu)}\right]m(\mu,H,T)\ \ \mu f(\mu)d\mu,\quad (19)$$

we express the conductivity in the form[14]

$$\sigma=c_0\frac{\xi_0+2\lambda_0\xi_1^2}{\xi_0^2-\xi_1^2},\quad (20)$$

in which $\lambda_0=2\sigma_m/c_0$ is the mean-free path in the pure matrix [Eqs. (24) and (25) below].

The magnetoresistance and the magnetoresistance ratio are, therefore,

$$\Delta\rho=\rho(H,T)-\rho(H_c,T)=-\frac{1}{c_0}\frac{\xi_1^2+2\lambda_0\xi_1^2\xi_0}{\xi_0+2\lambda_0\xi_1^2},\quad (21)$$

$$\mathrm{MR}=\frac{\Delta\rho}{\rho(H_c,T)}=-\frac{\xi_1^2+2\lambda_0\xi_1^2\xi_0}{\xi_0^2+2\lambda_0\xi_1^2\xi_0},\quad (22)$$

in which H_c is the coercive field. Note that the absolute value of the GMR amplitude is smaller than 1, because $\xi_0>\xi_1$.

Let us examine the two limits when $1\gg 2k_{+-}^{\mathrm{CPP}}(\mu)$ and when $1\ll 2k_{+-}^{\mathrm{CPP}}(\mu)$. In the small k_{+-}^{CPP} limit, $\xi_0\gg 2\lambda_0\xi_1^2$ and thus the total conductivity takes the form

$$\sigma=c_0\frac{\xi_0}{\xi_0^2-\xi_1^2}\quad (23)$$

which coincides with the expression of the conductivity in the theory of Zhang and Levy.

Comparing term by term Eqs. (6)–(11) in Ref. 4 with Eqs. (17)–(23), we determine the following relationships among the corresponding parameters:

$$c_0=\frac{ne^2k_F}{m\epsilon_F},\quad (24)$$

$$\frac{1}{\lambda_{nm}}=\frac{c_0}{2(1+2f)\sigma_m},\quad (25)$$

$$\frac{1+p_b^2}{\lambda_m}=\frac{c_0}{2(1+2f)}\frac{3}{2}\left(\frac{1}{\sigma_-}+\frac{1}{\sigma_+}\right),\quad (26)$$

$$\frac{(36\pi)^{1/3}(1+p_s^2)}{\lambda_s/a_0}=\frac{c_0}{2(1+2f)}I_s^{-1/3}\frac{3}{2}(r_-+r_+),\quad (27)$$

$$\frac{2p_b}{1+p_b^2}=\frac{1/\sigma_--1/\sigma_+}{1/\sigma_-+1/\sigma_+},\quad (28)$$

$$\frac{2p_s}{1+p_s^2}=\frac{r_--r_+}{r_-+r_+}.\quad (29)$$

The parameter α introduced in our previous paper[5] takes the form

$$\alpha=\frac{r_--r_+}{1/\sigma_--1/\sigma_+}.\quad (30)$$

Therefore, when $1\gg 2k_{+-}^{\mathrm{CPP}}(\mu)$, the magnetoresistance for granular alloys can be derived using the formalism for currents perpendicular to the plane (CPP) in layered structures. This is the CPP limit.

In the large k_{+-}^{CPP} limit, $\xi_1\approx 0$ and the total conductivity takes the form

$$\sigma=\frac{c_0}{\xi_0},\quad (31)$$

which does not depend on the directions of the magnetic moments. Therefore, the electrons avoid the high resistivity granules, the transport occurs mainly through the matrix, and the GMR vanishes. This is the CIP limit.

In the CPP limit, the granule conductivities are larger than the matrix conductivity and the granules are larger than a certain minimum size,

$$\text{CPP limit:}\quad \sigma_{+-}>2\sigma_m,\ \ V_\mu\gg V_{+-}^{\mathrm{CPP}}=(L_{+-}^{\mathrm{CPP}})^3=\left(\frac{2\sigma_m r_{+-}\sigma_{+-}I_s^{-1/3}}{\sigma_{+-}-2\sigma_m}\right)^3.\quad (32)$$

In the CIP limit, either the granule conductivities are smaller than the matrix conductivity ($\sigma_{+-}<2\sigma_m$) or the granules are smaller than a certain minimum size ($V_\mu\ll V_{+-}^{\mathrm{CPP}}$). Of course, L_{+-}^{CPP} are the relevant length scales.

Zhang and Levy attribute the initial increase of the GMR amplitude as a function of the annealing temperature to the superparamagnetic behavior of the smaller granules.[4] However, because of Eq. (32), this explanation is not consistent when the measurements are made at the blocking temperature of the smallest granules. The correct explanation is that the magnetoresistance drops when the alloy is close to the CIP limit.[6–8]

Camblong, Levy, and Zhang[11] set the general framework for a theory of electron transport in magnetic inhomogeneous media based on the real-space Kubo formula, and succeeded in obtaining the magnetoresistance for the CIP and CPP geometries in multilayers. Although restricting our approach to the formalism of classical electrodynamics, we extended the theory of transport in granular alloys beyond the CPP approach of Ref. 4 and the simplifying conditions of Refs. 6–8.

Measurements of the magnetization and magnetoresistance on melt-spun Co-Cu ribbons at high fields and low temperatures are being made to test our theory.

In conclusion, we developed the theory of giant magnetoresistance in granular alloys by taking into account the spin-dependent scattering within the magnetic granules and at their boundaries and the continuous distributions of granule sizes and directions of magnetization. The CPP model was recovered in the limit when the granule conductivities are larger than the matrix conductivity and the granule sizes are much larger than a certain length scale. When these conditions are not satisfied, the transport properties of the granular alloys derive from the crossover of the parallel and perpendicular current configurations.

APPENDIX

In fact, the electric fields and the current densities are the diagonal components of the bispinors $\boldsymbol{E}_{\beta}^{\alpha}$ and $\boldsymbol{j}_{\alpha}^{\beta}$ which are connected through the linear relationship (Appendix A of Ref. 11)

$$\boldsymbol{j}_{\alpha}^{\beta}(\boldsymbol{r})=\int \sigma_{\alpha\gamma}^{\beta\delta}(\boldsymbol{r},\boldsymbol{r}')\cdot\boldsymbol{E}_{\delta}^{\gamma}(\boldsymbol{r}')d^3r'. \tag{A1}$$

We assume that in the most symmetric configuration when the quantization axis is parallel to the magnetic moment Eq. (A1) reduces to the local form of Ohm's law,

$$\boldsymbol{j}_{+-}=\sigma_{+-}\boldsymbol{E}_{+-}, \tag{A2}$$

in which $\boldsymbol{j}_{+}\equiv\boldsymbol{j}_{+}^{+}$, $\boldsymbol{E}_{+}\equiv\boldsymbol{E}_{+}^{+}$, and $\sigma_{+}\equiv\sigma_{++}^{++}$, etc.

The change of the quantization axis from the magnetic moment direction to the field direction arises from the rotation characterized by the spherical angles (θ,φ) and the matrix

$$C=\begin{pmatrix}\cos(\theta/2) & \sin(\theta/2)e^{-i\varphi}\\ -\sin(\theta/2)e^{i\varphi} & \cos(\theta/2)\end{pmatrix}. \tag{A3}$$

The bispinors $\boldsymbol{s}=\boldsymbol{E},\ \boldsymbol{j}$ transform according to

$$\boldsymbol{s}'^{\alpha}_{\beta}=(C^{-1})^{\alpha}_{\gamma}\boldsymbol{s}^{\gamma}_{\delta}(C)^{\delta}_{\beta} \tag{A4}$$

and after simple algebraic operations take the form:

$$\boldsymbol{s}'=\begin{pmatrix}\boldsymbol{s}_{+}\cos^2(\theta/2)+\boldsymbol{s}_{-}\sin^2(\theta/2) & (\boldsymbol{s}_{+}-\boldsymbol{s}_{-})\sin(\theta/2)\cos(\theta/2)e^{-i\varphi}\\ (\boldsymbol{s}_{+}-\boldsymbol{s}_{-})\sin(\theta/2)\cos(\theta/2)e^{i\varphi} & \boldsymbol{s}_{+}\sin^2(\theta/2)+\boldsymbol{s}_{-}\cos^2\theta/2\end{pmatrix}. \tag{A5}$$

Therefore, the diagonal terms reduce to Eqs. (1) and (2) and the nondiagonal terms average out with the azimuthal integration.

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[13] A similar problem is solved, for example, by Gu *et al.* in Ref. 6, Sec. III. Our coefficients A_{+-} and D_{+-} given by Eqs. (14) and (15) reduce, after simple algebraic manipulations, to the corresponding coefficients given by their Eq. (18).

[14] The parameter c_0 is introduced for convenience. Note that it cancels out when Eqs. (17)–(19) are replaced in Eq. (20).