MAGNETORESISTANCE IN METALS WITH EMBEDDED MAGNETIC NANOCLUSTERS

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We propose a kinetic theory of transport in metals embedded with ferromagnetic nanoclusters, predicting a quadratic low-field magnetoresistance in an ensemble of clusters in thermodynamic equilibrium. We show that this effect is strongest when all the clusters have the same intrinsic easy-axis anisotropy.

Keywords: Mesoscopic systems; magnetic systems; giant magneto-resistance.

1. Introduction

Numerous applications of magnetoresistance (MR) phenomenon fuel a continuous search for new materials, including various hybrid systems. In this article, we propose a theory of high temperature MR in a normal metal embedded with ferromagnetic metallic nano-clusters (FmnC). In our model we assume that both the magnetisation and the electron exchange spin-splitting $J$ are homogeneous within each cluster (due to their small size, typically $10^3$ atoms per cluster) and we characterise the ensemble of FmnCs using the angular distribution of magnetic moments $m = \mu I$ of individual clusters, which depends on their intrinsic anisotropy, external magnetic field and temperature. This study is restricted to materials with a low cluster density and to high temperatures since we neglect correlations between clusters.

Due to band mismatch between underlying materials, each FmnC would generate a scalar potential well and have a different electron mass from that in the normal metal, in addition to a spin dependent (exchange) perturbation. An interplay between all above-mentioned factors makes electronic scattering from each individual FmnC spin-dependent and the resistance of a composite material sensitive to the mean degree of polarisation of clusters $\langle l_z \rangle$. The proposed theory predicts a quadratic MR effect, $\Delta(B) = \frac{R(B) - R(0)}{R(0)} \sim -B^2$ in the low field regime $\mu B/kT \ll 1$ and relates the form of MR to the magnetic field dependence of mean polarisation of FmnCs and its variance. The effect is strongest when all clusters in the ensemble have collinear easy-axis anisotropy, whereas it is suppressed by electron spin relaxation in ensembles with isotropic magnetic properties.
To describe the MR in a metal with FnnCs we study the quantum kinetic equation,
\[ \partial_t \hat{\rho} + \mathbf{v} \cdot \nabla \hat{\rho} + e(\mathbf{E} \cdot \mathbf{v}) \partial_v \hat{\rho} - \frac{\omega_B}{2} [\hat{\sigma}_3, \hat{\rho}] = I_0[\hat{\rho}] + \langle I[\hat{\rho}] \rangle \]
for the electron spin density matrix \( \hat{\rho} = \rho_0 + \sum_i \rho_i \hat{\sigma}_i \) approximated by \( \rho_0(\mathbf{p}) = \tilde{\rho}_0(p) + \frac{\mathbf{p} \cdot \mathbf{n}_0(p)}{p} \) where \( \sigma = (\hat{\sigma}_1, \hat{\sigma}_2, \hat{\sigma}_3) \) are Pauli matrices, \( \omega_B = \frac{BeH}{mc} \) is the electron spin precession frequency in normal metal and \( \tilde{\rho} \) is the isotropic part of the electron distribution in momentum space. All details of the electron spin and phase evolution inside clusters are incorporated into the collision integral \( I[\hat{\rho}] \), whereas momentum relaxation due to non-magnetic impurities and phonons in the normal metal is taken into account by \( I_0[\hat{\rho}] = [\hat{\rho}(\mathbf{p}) - \tilde{\rho}(p)] \tau^{-1} \).

One FnnC produces the following scattering matrix,
\[ \hat{S} = a + b(\sigma \cdot 1); \quad a = \frac{S^+ + S^-}{2}, \quad b = \frac{S^- - S^+}{2}, \]
(1)
where \( S^+_{pp'} \) and \( S^-_{pp'} \) characterise the angular dependence of scattering matrices of electrons with spin parallel and antiparallel to the cluster polarisation \( \mathbf{I} [\mathbf{p}, \mathbf{p}' \text{ are electron momenta before and after scattering}]. \) The collision integral taking into account a group of collinearly polarised clusters with the concentration \( n(I) \) has the form
\[ I_1[\hat{\rho}] = \frac{n(I) \hbar^2}{4mp} \int d^3p' \left\{ \hat{S}_{pp'} \hat{\rho}_{pp'} \hat{S}^\dagger_{pp'} - \frac{1}{2} (\hat{S}_{pp'} \hat{S}^\dagger_{pp'} \hat{\rho} + \hat{\rho} \hat{S}_{pp'} \hat{S}^\dagger_{pp'}) \right\} \]
(2)
where \( v \) is Fermi velocity. This expression incorporates the kinetics of both electron scattering and spin precession, in particular since \( S \) matrix takes into account spin-dependent forward scattering. Due to the angular distribution of cluster polarisations, the ensemble-averaged collision integral, \( \langle I[\hat{\rho}] \rangle \) would also describe spin relaxation of electrons. All of the above-mentioned three generic elements of electron spin kinetics would be present even if the distribution of cluster magnetic moments is axially symmetric about the direction of an external magnetic field \( \mathbf{B} = Be_z \).

Therefore, in the following analysis we assume that \( \langle l_x \rangle = \langle l_y \rangle = 0 \) and the tensor \( \langle l_z \rangle \) is diagonal with \( \langle l^2_z \rangle = \langle l^2_y \rangle = 1 - \langle l^2_z \rangle \). The resulting ensemble-averaged collision integral reads as
\[ \langle I[\hat{\rho}] \rangle = \frac{n_c \hbar^2}{4mp} \int d^3p' \left\{ (ab^* + a^*b)(\rho'_3 - \rho_3) \langle l_z \rangle + (|a|^2 + |b|^2)(\rho'_0 - \rho_0) \right. \]
\[ + (|a|^2 + |b|^2) \sum_i \hat{\sigma}_i (\rho'_i - \rho_i) - 2|b|^2 \sum_i \hat{\sigma}_i \rho'_i \]
\[ + i(a^*b - ab^*)(\rho'_3 \langle l_z \rangle \hat{\sigma}_1 - \rho_1 \langle l_z \rangle \hat{\sigma}_2) \]
\[ + (ab^* + a^*b)(\rho'_0 - \rho_0) \langle l_z \rangle \hat{\sigma}_3 + 2|b|^2 \sum_i \rho'_i \langle l^2_z \rangle \hat{\sigma}_i \left. \right\} \]
where density matrix \( \hat{\rho} \) was decomposed into a singlet \( (\rho_0) \) and triplet \( (\rho_i) \) parts [here \( \rho = \rho(\mathbf{p}, \mathbf{p}') = \rho(\mathbf{p}') \)], which decouples the kinetic equation into two groups.
The first group describes the evolution of only diagonal density matrix components and involves momentum ($\tau^{-1}_{p\pm}$) and spin ($\tau^{-1}_{sn}$) relaxation rates,

\[ \partial_t \rho_0 + \frac{v}{3} \nabla \cdot \eta_0 = 0 \]
\[ \partial_t \rho_3 + \frac{v}{3} \nabla \cdot \eta_3 = -(1 - \langle l_z^2 \rangle) \tau^{-1}_{s0} \rho_3 \]
\[ v \nabla \rho_0 + evE \partial_z \rho_T(z) = -\tau^{-1}_{p+} \eta_0 - \tau^{-1}_{p-} \langle l_z \rangle \eta_3 \]
\[ v \nabla \rho_3 = -\tau^{-1}_{p+} \langle l_z \rangle \eta_0 - \tau^{-1}_{p+} (1 - \langle l_z^2 \rangle)) \eta_3, \]
\[ \tau^{-1}_{p+} = \frac{n_e h^2 \pi}{4 m_p} \int d\theta \sin \theta (|S^+|^2 + |S^-|^2) (1 - \cos \theta) + \frac{1}{\tau} \]
\[ \tau^{-1}_{p-} = \frac{n_e h^2 \pi}{4 m_p} \int d\theta \sin \theta (|S^+|^2 - |S^-|^2) (1 - \cos \theta), \]
\[ \tau^{-1}_{sn} = \frac{n_e h^2 \pi}{4 m_p} \int d\theta \sin \theta |S^+ - S^-|^2 \cos^n \theta. \]

The second group,

\[ \partial_t \zeta + \frac{v}{3} \nabla \cdot \zeta = -(\langle l_z^2 \rangle \tau^{-1}_{s0} \zeta - \omega \zeta \]
\[ v \nabla \zeta = -\langle \tau^{-1}_{p+} (1 - \langle l_z^2 \rangle) \rangle \eta_0 - \langle \tau^{-1}_{p+} + \langle l_z^2 \rangle \rangle \eta_3, \]

where $\zeta = \rho_1 + i \rho_2$ and $\eta = n_1 + in_2$, takes into account an average of electron spin precession in the $(xy)$-plane with frequency

\[ \omega = \langle l_z \rangle \frac{n_e h^2 \pi}{2 m_p} \int \text{Im}(S^+ S^-) \sin \theta d\theta + \omega_B. \]

Equation (3) describes the kinetics of charge ($\rho_0$) and spin ($\rho_3$) densities in a macroscopic sample. Without any electric bias this would be diffusion described by equations

\[ \left\{ \begin{array}{l}
 \partial_t \rho_0 = D_\Delta \nabla^2 \rho_0 - D_{\nabla^2} \nabla^2 \rho_3 \\
 \partial_t \rho_3 = D_\Delta \nabla^2 \rho_3 - D_{\nabla^2} \nabla^2 \rho_0 - (1 - \langle l_z^2 \rangle) \tau^{-1}_{s0} \rho_3 
\end{array} \right., \]

\[ \text{Eq. (4) is a pair of diffusion equations for in plane kinetic} \]

\[ \partial_t \zeta = D_{\nabla^2} \nabla^2 \zeta - iD'_{\nabla^2} \nabla^2 \zeta - (1 - \langle l_z^2 \rangle) \tau^{-1}_{s0} \zeta - \omega \zeta, \]

\[ D_{\nabla^2} = \frac{v^2}{3} \frac{(\tau^{-1}_{p+} + (1 - \langle l_z^2 \rangle) \tau^{-1}_{s1})}{(\tau^{-1}_{p+} + (1 - \langle l_z^2 \rangle) \tau^{-1}_{s1})^2 + (l_3 \omega)^2} \]

\[ D'_{\nabla^2} = \frac{v^2}{3} \frac{(l_3 \omega)}{(\tau^{-1}_{p+} + (1 - \langle l_z^2 \rangle) \tau^{-1}_{s1})^2 + (l_3 \omega)^2}. \]
\[ D_e = \frac{v^2}{3} \frac{(\tau_{p+}^{-1} + \tau_{s1}^{-1} (1 - \langle l_z^2 \rangle))}{\tau_{p+}^{-1} (\tau_{p+}^{-1} + \tau_{s1}^{-1} (1 - \langle l_z^2 \rangle)) - \tau_{p-}^{-2} \langle l_z \rangle^2}, \]

\[ D_s = \frac{v^2}{3} \frac{\tau_{p+}^{-1}}{\tau_{p+}^{-1} (\tau_{p+}^{-1} + \tau_{s1}^{-1} (1 - \langle l_z^2 \rangle)) - \tau_{p-}^{-2} \langle l_z \rangle^2}, \]

\[ D_\Delta = \frac{v^2}{3} \frac{(\langle l_z \rangle \tau_{p+}^{-1})}{\tau_{p+}^{-1} (\tau_{p+}^{-1} + \tau_{s1}^{-1} (1 - \langle l_z^2 \rangle)) - \tau_{p-}^{-2} \langle l_z \rangle^2}. \]

A homogeneous solution of Eq. (3) in the presence of electric bias produces the current-field relation for both charge and spin components \( j_e = \sigma_e E \) and \( j_s = \sigma_s E \), which determines electric \((e)\) and spin \((s)\) conductivities,

\[ \sigma_e = e^2 \nu (\varepsilon_F) D_e, \quad \sigma_s = -(h/2)e\nu (\varepsilon_F) D_\Delta. \quad (6) \]

A magnetic field dependence of average cluster polarisation \( \langle l_z \rangle \) and mean square \( \langle l_z^2 \rangle \) generates a finite magneto-resistance (MR) of a composite material,

\[ \Delta(B) = \frac{\sigma_e(0)}{\sigma_e(B)} - 1. \quad (7) \]

The resistance change which can be achieved across an “infinite” magnetic field interval (providing a complete uniaxial polarisation of all clusters) is given by

\[ \Delta_{\text{max}} = \left( \frac{\tau_{p+}}{\tau_{p-}} \right)^2, \]

whereas the entire MR curve can be described by

\[ \Delta(B) = -\frac{\langle l_z \rangle^2 \Delta_{\text{max}}}{1 + (1 - \langle l_z^2 \rangle)X}, \quad \text{where } X = \frac{\tau_{p-}}{\tau_{p+} + \tau_{s1}}. \quad (8) \]

The values of \( \langle l_z \rangle \) and \( \langle l_z^2 \rangle \) in Eq. (8) vary with a magnetic field and depend on magnetic anisotropy of clusters. The parameter \( X \) is determined by the material composition and cluster size. For large spherical clusters \((pr_{cl} \gg h)\) all relaxation rates are proportional to the clusters’ geometrical cross-section, \( \tau_\alpha^{-1} = \frac{m}{m_{NP}^+} \pi (pr_{cl})^2 A_\alpha \), \((\alpha = p+, p-, s0, s1)\). Table 1 illustrates values of these parameters for various ratios between the exchange \( J \) and Fermi energy \( E_F \) in the ferromagnet and the ratios

<table>
<thead>
<tr>
<th>( J/E_F )</th>
<th>( m_F/m_N )</th>
<th>( A_{p+} )</th>
<th>( A_{p-} )</th>
<th>( A_{s0} )</th>
<th>( A_{s1} )</th>
<th>( X )</th>
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<td>1</td>
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<td>0.7</td>
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</table>
Magnetoresistance in Metals with Embedded Magnetic Nanoclusters

...m_F/m_N between band masses in the normal and ferromagnetic metals. Here, we use a simplifying model with parabolic dispersion in both materials with equal densities of conduction band electrons. The latter constraint determines a spin-independent band mismatch \( U = E_F^N - E_F^F \) at the interface which is taken into account as a scattering potential. The examples in Table 1 also assume that \( \tau \rightarrow \infty \) in normal metal.

Below, we compare the magnetic field dependence \( \Delta(B) \) for three ensembles of clusters: (a) intrinsically isotropic FmnCs; (b) identical clusters with colinear easy axes aligned with the external magnetic field; (c) ensemble of clusters with randomly oriented easy axes. In all of these three cases we assume the same material composition, size of clusters, surrounding medium (i.e. fixed values of \( \tau_{p \pm}, \tau_{sn} \) and \( X \)), and thermal equilibrium of angular distribution of clusters. These three situations will be identified with the following form of free energy of magnetic subsystems,

\[
F = -\mu \mathbf{B} \cdot \mathbf{l} + F_0 = \begin{cases} 
-\mu B l_z 
& (a) \\
-\mu B l_z - \alpha l_z^2 
& (b) \\
-\mu B l_z - \alpha (1 \cdot l_0)^2 
& (c)
\end{cases}
\]

where \( l_0 \) determines the direction of single cluster easy axis. In cases (a) and (b) the values of \( \langle l_z \rangle \equiv l_z \) and \( \langle l_z^2 \rangle \equiv l_z^2 \) are determined by the thermal average

\[
\tilde{A} = N^{-1} \int d\Omega_4 A e^{-\frac{F_0}{kT}}, \quad N = \int d\Omega_4 e^{-\frac{F_0}{kT}},
\]

where \( T \) is temperature. For ensemble (c) with isotropic distribution of easy axes, an additional averaging over directions of vector \( l_0 \) in Eq. (9c) is needed \( \langle l_z \rangle = \langle \langle l_z \rangle \rangle \) and \( \langle l_z^2 \rangle = \langle \langle l_z^2 \rangle \rangle \), where \( \langle \langle \cdots \rangle \rangle = \int d\Omega_4 h_{\langle \cdots \rangle} / 4\pi \).

For intrinsically isotropic clusters, ensemble type (a),

\[
\langle l_z \rangle_a = (x \coth x - 1) / x, \quad x = B \mu / kT, \\
\langle l_z^2 \rangle_a = (x^2 - 2x \coth x + 2) / x^2.
\]

The resulting form of MR is illustrated in Fig. 1 by the curve (a). The influence of intrinsic easy-axis anisotropy of clusters on MR depends on the temperature regime and the type of cluster ensemble (b) or (c). At high temperatures \( kT > \alpha \), the existence of an anisotropy axis common for all clusters results in a small correction to \( \langle l_z \rangle \), \( \langle l_z^2 \rangle \) in ensemble (b),

\[
\langle l_z \rangle_b = \langle l_z \rangle_a + \frac{\alpha}{kT} L_1^b(x), \quad \langle l_z^2 \rangle_b = \langle l_z^2 \rangle_a + \frac{\alpha}{kT} L_2^b(x),
\]

thus leading to small corrections to \( \Delta(B) \). Here \( L_1^b(x) = [2x^2(\coth^2 x - 1) + 2x \coth x - 4] / x^3 \) and \( L_2^b(x) = [4x^2(2 - \coth^2 x) - 16x \coth x + 20] / x^4 \). In ensemble (c), which has an isotropic distribution of magnetic moments at \( B = 0 \), such a correction is even smaller and appear only in the second order in \( \alpha / kT \leq 1 \).
At low temperatures, $kT < \alpha$, $\Delta(B)$ was studied numerically. The obtained results for ensembles (b) and (c) are compared in Fig. 1 with the MR in ensemble (a). Figure 1 shows that a pronounced easy-axis anisotropy enhances the low-field MR effect, whereas an isotropic distribution of easy axes suppresses MR via spin relaxation. The inset to Fig. 1 demonstrates the low-field behaviour in all three cases, which complies with the asymptotic expansion of $\Delta(B)$ in Eq. (8) in powers of $\mu B/kT \ll 1$. Since at small fields $e^{\mu B l_z/kT - F_0/kT} \approx e^{-F_0/kT}(1 + \mu B/kT l_z)$,

$$\langle l_z \rangle \approx \frac{\mu B}{kT} \int d\Omega l_z^2 \exp(-F_0/kT) \int d\Omega \exp(-F_0/kT) = \frac{\mu B}{kT} \langle l_z^2 \rangle_{B=0},$$

hence the low-field MR is quadratic,

$$\Delta(B) \approx -\left(\frac{\mu B}{kT}\right)^2 \frac{\Delta_{\text{max}}(l_z^2)_{B=0} \Delta_{\text{max}}(l_z^2)_{B=0}}{1 + (1 - \langle l_z^2 \rangle_{B=0})X}.$$ 

Due to isotropy of FmnC ensembles (a) and (c) at $B = 0$, $\langle l_z l_j \rangle_{B=0} = \frac{1}{3} \delta_{ij}$ and $\langle l_z^2 \rangle_{a,B=0} = \langle l_z^2 \rangle_{c,B=0} = \frac{1}{3}$, whereas

$$\langle l_z^2 \rangle_{b,B=0} = f(\alpha/kT), \quad f(y) = \frac{ye^{y}y^{-1/2}}{\text{erf}(y^{1/2})} - \frac{1}{y}.$$ 

The low-field MR in each of those three cases is

$$\frac{\Delta(B)}{\Delta_{\text{max}}} = -\left(\frac{\mu B}{kT}\right)^2 \begin{cases} (9 + 6X)^{-1} & \text{(a, c)} \\ \frac{f^2(\alpha/kT)}{1 + [1 - f(\alpha/kT)]X} & \text{(b)} \end{cases}.$$ 

Fig. 1. $\Delta(B)$ for different cluster subsystems (a) intrinsically isotropic clusters; (b) identical clusters with colinear easy axis in direction of external magnetic field; (c) ensemble of clusters with randomly oriented easy axis; $kT = 0.1$, $\alpha = 1$. The used parameters correspond to the first row of Table 1, $\tau \to \infty$ and $X = 27.5.$
In summary, we propose the theory of classical magnetoresistance in a normal metal with embedded ferromagnetic nanoclusters. Our analysis shows that in a thermodynamic equilibrium the MR effect is largest when all clusters have the same intrinsic easy-axis anisotropy. Thus it may be advantageous for applications to engineer anisotropy by growing the elongated clusters as sketched in Fig. 1 — even despite the inconvenience which may be caused by hysteresis in their magnetisation dynamics. As spin-orbit coupling would cause suppression of the MR by producing spin relaxation, i.e. an addition to $\tau_{\text{so}}^{-1}$, so the effect should be more pronounced in light metals, for instance Al.

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References
