Giant magnetothermopower and magnetoresistance in metals with embedded ferromagnetic nanoclusters

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We show that in granular normal-ferromagnetic metals the giant magnetothermopower is related to the giant magnetoresistance as it is a result of the interplay between the spin-dependent elastic scattering (responsible for magnetoresistance effect) and the inelastic spin mixing scattering on magnetic clusters. For a small change of resistance of sample in an applied magnetic field the variation of the thermopower is connected linearly with the giant magnetoresistance and both are proportional to the square of the sample magnetization. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2409282]

After the discovery of giant magnetoresistance (GMR) in magnetic multilayers,[1] a lot of attention has been attracted to various combinations of normal and ferromagnetic materials driven by possible device applications.[2] Later the same phenomenon was uncovered in granular media consisting of single domain ferromagnetic particles in a nonmagnetic noble matrix.[3,4] GMR was naturally explained in terms of spin-dependent electron transport suggested by a polarization of the magnetic component in an applied field.[5,6] Later, thermoelectric power in these systems was also found to have a strong dependence on the magnetic field and a correlation between GMR and magnetothermopower (MTP) has been registered[7–12] and attributed to the energy dependent density of states in a weakly ferromagnetic metal with a complex band structure.

In this article we show that a strong magnetic field dependent contribution to the thermopower coefficient, $\delta c = c(B) - c(0)$, rises routinely in kinetic theory from an interplay between the inelastic contribution towards the momentum relaxation of electrons caused by spin-flip processes involving polarized magnetic scatterers and a spin-dependent elastic scattering responsible for the GMR effect. The magnetic field dependence of δc tracks the magnetic field dependence of GMR,

$$\delta c(B) = \frac{k}{e}\frac{\tau_s^{-1}}{\tau_\uparrow^{-1} - \tau_\downarrow^{-1}}\Delta(B), \tag{1}$$

where the proportionality coefficient is the ratio between the spin-flip transfer rate τ_s^{-1} and the relative difference between the inverse mean free path time $\tau_{\uparrow(\downarrow)}^{-1}$ of electrons with spins parallel (antiparallel) to the cluster polarization axis, k is the Boltzmann constant, and e is the electron charge. In a metal with ferromagnetic clusters[5] or magnetic impurities[13] with a large spin $S \gg 1$, the magnetoresistance, $\Delta \equiv R(B)/R(0) - 1$,

$$\Delta(B) = -\left(\frac{\tau_\uparrow - \tau_\downarrow}{\tau_\uparrow + \tau_\downarrow}\right)^2 \langle l_z \rangle^2, \tag{2}$$

reflects the degree of polarization, $\langle l_z \rangle = \coth(y) - y^{-1}$, $y = \mu BS/kT$, of the magnetic subsystem by a magnetic field $\mathbf{B} = \mathbf{n}_z B$. The strength of the MR effect depends on the relative difference between the mean free path times $\tau_\uparrow$ and $\tau_\downarrow$. Here all relaxation rates are formally determined for a system with fully polarized clusters and equal density of states γ for electrons with spin-↑ and spin-↓.

The diffusive nature of transport properties in granular normal-ferromagnetic heterogeneous metals was confirmed to agree with the Wiedemann-Franz law. For instance, in a Co–Ag compound it holds throughout the temperature range of 2–300 K.[9] When two parts of a sample polarized by an external magnetic field are kept at different temperatures, $T_1 > T_2$, the heat flux which equilibrates the thermal conditions on the opposite sides has to be accompanied by a transport of magnetization, i.e., spin current. Locally, the process of such an equilibration involves the spin transfer from clusters to conduction electrons via spin-flip scattering processes. Then, the magnetization is carried by electrons diffusing between sample areas kept at different temperatures, thus leading to a spin current. Due to the difference between mean free path times (thus, mobilities) of "up" and "down" spin carriers, the spin current also drags a charge current. In an open circuit, this generates a thermopower.

A microscopic justification of this phenomenological argument is supported by the following analysis of the electron-hole symmetry breaking in such a system, which represents a necessary attribute of thermoelectric effect. In normal metals diffusive thermopower is caused by the energy dependence of the density of states near the Fermi energy, so that $c \sim (\pi^2/3)(k/e)(kT/\epsilon_F)$. In a magnetic material electron-hole asymmetry is created in alternative way—via the formation of energy and spin dependence of quasiparticle kinetic properties, such as its scattering rate.[14,15]

We model ferromagnetic nanoclusters (FmnC's) using the Hamiltonian[16]

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$$-\mu BS^z + \int \psi^\dagger(\mathbf{r}) \sum_i (U + J\boldsymbol{\sigma}\cdot\hat{\mathbf{S}}_i)\delta(\mathbf{r}-\mathbf{r}_i)\psi(\mathbf{r})d^3\mathbf{r}.$$

Here, U takes into account the FmnC charge and mismatch in band structures between normal and magnetic metals, whereas J is the exchange interaction, σ is the spin of the electron, B is the external magnetic field, and μ is the Bohr magneton, so that μS is the magnetic moment of a cluster. The GMR analysis in materials with large spin clusters, $S \gg 1$, can be done using a macrospin model,[5] where the operator $\mathbf{S}$ is replaced by $S\mathbf{l}$ (here, $\mathbf{l}$ is a unit vector in the direction of the polarization of an individual cluster). In contrast, the analysis of the MTP and tracking a kinetically generated electron-hole asymmetry necessitates taking into account the quantum nature of the cluster spin via the electron spin-flip process.

To find the transport coefficients we solve a steady state kinetic equation,

$$[\mathbf{v}\nabla - e\mathbf{E}\partial_{\mathbf{p}}]\rho_\pm = \langle I_\pm[\rho_+,\rho_-]\rangle, \tag{3}$$

where the collision integrals $I_\pm$ describe the balance in the distribution functions $\rho_\pm$ of electron with spin-↑ and spin-↓ formed via scattering from a group of FmnC's with given S_z, and we assume the paramagnetic state of the system,[17] thus average over the thermal distribution of initial state of cluster.

$$I_\pm[\rho] = \frac{2\pi n_c}{\hbar}\int \frac{d^3\mathbf{p}'}{h^3}[U^2 + S_z^2J^2 \pm 2UJS_z](\rho'_\pm - \rho_\pm)\delta(\epsilon_{\mathbf{p}} - \epsilon_{\mathbf{p}'}) + J^2[(S^2 - S_z^2 + S \pm S_z)\rho'_\mp(1-\rho_\pm) - (S^2 - S_z^2 + S \mp S_z) \times \rho_\pm(1-\rho'_\mp)]\delta(\epsilon_{\mathbf{p}'} - \epsilon_{\mathbf{p}} \mp \mu B). \tag{4}$$

In the above equation the first term describes elastic spin conserving processes ↑(↓)→↑(↓), whereas the second term describes inelastic spin-flip processes ↑(↓)→↓(↑), where scattering amplitudes are taken in the Bohr approximation. We restrict this study to a temperature higher than the Kondo temperature. By inspection of the second term, one can notice that the probabilities of electrons ↓→↑ and ↑→↓ spin-flip scattering processes from the same cluster slightly differ, so that in a system with a preferential polarization of FmnC's along an external magnetic field, this leads to the same behavior of electron momentum relaxation rate.

In the linear response approximation, temperature gradient and electric field are treated as small perturbations. Thus we expand the electron density function into angular harmonics in the momentum space, $\rho_\pm \approx \rho_T + \boldsymbol{\rho}_\pm\cdot\mathbf{p}/p$ (here, $\rho_T(\epsilon) = [e^{\epsilon/kT}+1]^{-1}$) and find that

$$\left(e\mathbf{E} - \frac{\epsilon-\epsilon_F}{T}\nabla T\right)v_F\partial_\epsilon\rho_T = -\frac{2\pi n_c\gamma}{\hbar}[U^2 + J^2S(S+1) \pm 2UJS\langle l_z\rangle \pm J^2S\langle l_z\rangle(2\rho_T(\epsilon \pm \mu B) \mp 1)]\boldsymbol{\rho}_\pm, \tag{5}$$

where the factor in front of $\boldsymbol{\rho}_\pm$ is the energy dependent momentum relaxation rate in each of two spin channels.[16] Since the inelastic part is a Fermi function shifted by a finite value (the Zeeman energy) from the Fermi level, Mott's formula cannot be used to calculate the thermopower coefficient[18] straight away. Evaluation of electrical current $\mathbf{j} = (e/3)\int d\epsilon\gamma(\rho_+ + \rho_-) = \sigma\mathbf{E} + \varkappa\nabla T$ for the limit of large spin from solution of Eq. (5) yields

$$\sigma = \frac{4e^2v_F^2\gamma}{3}\frac{\tau_\uparrow^{-1} + \tau_\downarrow^{-1}}{(\tau_\uparrow^{-1} + \tau_\downarrow^{-1})^2 - (\tau_\uparrow^{-1} - \tau_\downarrow^{-1})^2\langle l_z\rangle^2}, \tag{6}$$

$$\varkappa = \frac{8ev_F^2\gamma k}{3}\frac{(\tau_\uparrow^{-2} - \tau_\downarrow^{-2})\tau_s^{-1}\langle l_z\rangle^2 f(\mu B/kT)}{[(\tau_\uparrow^{-1} + \tau_\downarrow^{-1})^2 - (\tau_\uparrow^{-1} - \tau_\downarrow^{-1})^2\langle l_z\rangle^2]^2}. \tag{7}$$

Here, the factor $f(x) = x^2e^x/(e^x-1)^2$, $x = \mu B/kT$, takes into account the availability of thermal electrons to transfer energy $\epsilon - \epsilon' = \mu B$ when impurity changes the spin state and is specific for the case $S \gg 1$.[19] In the above mean free path scattering times are defined in a completely polarized sample,

$$\tau_{\uparrow(\downarrow)}^{-1} = \frac{2\pi n_c\gamma}{\hbar}[U^2 + J^2S(S+1) \pm 2UJS], \tag{8}$$

$$\tau_s^{-1} = \frac{4\pi n_c\gamma}{\hbar}J^2S.$$

Having expressed both the conductivity σ (which determines the MR in a metal embedded with FmnC's) and $\varkappa$ in terms of the degree of polarization of FmnCs $\langle l_z\rangle$, we can relate the MTP, $\delta c = -\sigma/\varkappa$, caused by the electron spin-flip scattering resulting in the e-h asymmetry to the MR coefficient,

$$\delta c(B) = \frac{k}{e}\frac{\tau_s^{-1}}{\tau_\uparrow^{-1} - \tau_\downarrow^{-1}}\frac{\Delta(B)}{1+\Delta(B)}f\left(\frac{\mu B}{kT}\right). \tag{9}$$

The magnetic field dependence of MTP implicit in the above result has two scales in it. First, at a low magnetic field where the polarization of clusters develops, the MTP is proportional to the MR and saturates together with $\Delta(B)$ at the field, where $B_* \sim kT/S\mu$ and $f(\mu B/kT \ll 1) \approx 1$. Then, at a much higher field range, $B_1 \sim kT/\mu$, the MTP gets suppressed and dies away. Also, the MR effect in the experimentally studied metallic structures embedded with ferromagnetic clusters is weak, $\Delta \lesssim 0.1$, so that in the field range $B_1 < kT/\mu$, we can relate the GMR and MTP effects to each other as in Eq. (1).

Bending of electron trajectories by a magnetic field generates the Hall contribution to the electric current. Thus the

conductivities σ and $\varkappa$ in the definition of $\mathbf{j}$ before Eq. (6) become tensors. The off-diagonal components of $\hat{\sigma}$ describe the Hall effect, $R_H=E_y/Bj_x$. Its dependence on the degree of polarization $\langle l_z\rangle$ is similar to that of the diagonal component of resistivity in Eq. (2),

$$R_H=\frac{3}{2v_F^2\gamma emc}[1-\Delta(B)], \tag{10}$$

where c is the velocity of light. The off-diagonal components of $\varkappa$ describe the Nernst effect, $N=E_y/B\partial_x T$. Its dependence on the degree of polarization is similar to that of the thermopower coefficient in Eq. (9),

$$N=\frac{8k}{mc}\frac{(\tau_\uparrow^{-1}-\tau_\uparrow^{-1})\tau_s^{-1}\langle l_z\rangle^2 f(\mu B/kT)}{(\tau_\uparrow^{-1}+\tau_\uparrow^{-1})^3[1+\Delta(B)]^2}. \tag{11}$$

In both results we assumed $\Omega(\tau_\uparrow^{-1}+\tau_\downarrow^{-1})^{-1}\ll 1$,[20] $\Omega=eB/mc$ is a cyclotron frequency.

In conclusion, we demonstrated that in the diffusive regime giant MTP is the result of interplay between an electron-hole asymmetric contribution towards the momentum relaxation of carriers in a metal caused by spin-flip processes involving polarized magnetic scatterers and a spin-dependent elastic scattering responsible for the GMR effect. In this case the giant MTP is proportional to the GMR for small values of GMR which gives an interpretation of experiments in Refs. 7–12 and both are proportional to the square of the sample magnetization.

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[19] For a metal with spin-$\frac{1}{2}$ impurities, their polarization and MR develop over the same range of magnetic field as the field suppressing the electron spin-flip scattering. Therefore, in a metal with spin-$\frac{1}{2}$ Anderson impurites, magnetothermopower would display a sharp nonmonotonic field dependence with a maximum effect at $B\approx 2\mu B/kT$, $\delta c(B)=-(k/e)(\tau_s^{-1}/\tau_-^{-1})\times[\Delta(B)/\cosh^2(\mu B/2kT)]$.
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