Thermally excited spin-current in metals with embedded ferromagnetic nanoclusters

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We show that a thermally excited spin-current naturally appears in metals with embedded ferromagnetic nanoclusters. When such materials are subjected to a magnetic field, a spin current can be naturally generated by a temperature gradient across the sample as a signature of electron-hole symmetry breaking in a metal due to the electron spin-flip scattering from polarised magnetic moments. Such a spin current can be observed via a giant magneto-thermopower which tracks the polarisation state of the magnetic subsystem and is proportional to the magnetoresistance. Our theory explains the recent experiment on Co clusters in copper by S. Serrano-Guisan et al [Nature Materials AOP, doi:10.1038/nmat1713 (2006)].

Theoretical studies of spin-currents in metals and semiconductors have recently been fuelled by the search for materials to develop spintronic devices [1]. Among the materials studied of particular interest had been composite ferromagnetic and nonmagnetic metallic systems, such as metallic multilayers [2] and metals with embedded ferromagnetic nanoclusters [3], where the electronic transport strongly depends on the electron spin leading to the effect of the giant magnetoresistance (MR).

In metals with ferromagnetic nanoclusters [3, 4] or magnetic impurities [5] with a large spin, s ≫ 1, the magnetoresistance reflects the degree of polarisation, ⟨lz⟩ = coth (y) − y−1, y = μBs/kT of a magnetic sub-system by a magnetic field B = nxB, where

\[\Delta(B) = \frac{R(B)}{R(0)} - 1 = \frac{⟨\tau↑ − \tau↓⟩^2}{4τ^2} ⟨lz⟩^2.\]

The MR effect develops across the magnetic field range \(B_{MR} \sim kT/\mu s\), and its strength depends on the relative difference between the mean free path time \(\tau↑(\downarrow)\) of electrons with spins parallel(antiparallel) to the cluster polarisation axis. Here, \(\tau↑(\downarrow)\) is defined as the mean free path in a metal with magnetic clusters whose spin polarisations are fully up(down). The Fermi density of states per spin, \(\gamma\), is identical for both electron spin orientations, and \(τ = \frac{1}{2} (τ↑ + τ↓)\) is a characteristic mean free path time, while \(μ\) is the Bohr magneton.

In this Letter we show that a thermally excited spin-current naturally appears in metals with embedded ferromagnetic nanoclusters (FmmC’s) when subjected to a magnetic field and temperature gradient. This effect appears through the manifestation of electron-hole symmetry breaking via electron spin-flip scattering from polarised magnetic moments. When two parts of a metallic sample with embedded FmmC’s are held at different temperatures \(T_1 > T_2\), the thermally equilibrating heat flux must be accompanied by a transport of magnetisation [2, 3, 4], i.e., a spin current, \(j_s\). This spin current acts to equilibrate the cluster polarisation on opposing sides of the temperature gradient. Locally, the equilibration process requires spin transfer from clusters to conduction electrons via spin-flip scattering at a rate \(τ_s^{-1}\), which is the inverse mean free path time of the spin-flip scattering process (formally defined latter, Eq.(1)). After a single spin-flip scattering event, the scattered electron will carry the transferred magnetisation while diffusing (with diffusion coefficient \(D\)) between different parts of the sample held at different temperatures, thus leading to a spin-current,

\[j_s = β∇T, \quad β \approx \frac{1}{2} \frac{h}{k} \gamma D \frac{τ_f}{τ_s} ⟨lz⟩.\] (1)

Here \(f = \frac{x^2e^x}{(e^x-1)^2}\) and \(x = \mu B/kT\) show that the spin current will persist up to a value of the magnetic field \(B_s \sim kT/\mu\), which is much larger than the typical field value at which MR develops, \(B_{MR} \ll B_s\). Additionally, due to the difference in the mean free path times of “up” and “down” spin carriers, \(⟨\tau↑ − \tau↓⟩⟨lz⟩\), the spin current ‘drags’ a charge current, \(j \propto x∇T\). In an open circuit, this generates a thermopower, \(V_{12} \propto c(B)[T_1−T_2]\), with a strong magnetic field dependence resembling (for \(B < kT/\mu\)) that of MR,

\[\Xi(B) ≡ c(B) − c(0) = \frac{k}{\epsilon} \frac{τ_s^2 f}{τ_s(τ↓ − τ↑)} Δ(B).\] (2)

A microscopic justification of the above-presented phenomenological argument is supported by the following analysis of the electron-hole asymmetry in a composite metal with ferromagnetic components. In normal metals, the electron-hole asymmetry leading to the thermopower is caused by the energy dependence of the density of states of electrons near the Fermi level. In materials containing a polarised magnetic subsystem, the electron-hole asymmetry is created in an alternative way — via the energy and spin dependence of a quasi-particle scattering rate [4]. In the system we discuss in this Letter, the formation of kinetic electron-hole asymmetry can be illustrated using Kondo-type model [11] which treats a FmmC as an impurity with a large spin, \(s\), and contains all the necessary ingredients to describe the MR effect simultaneously with the generation of spin-current and magneto-thermopower.
Thus, we model the FmnC’s by the Hamiltonian
\[ -\mu B\tilde{s}_z + \int \psi^\dagger(\mathbf{r}) \sum_i \left(U + J\mathbf{\delta} \cdot \mathbf{\hat{s}}(\mathbf{r} - \mathbf{r}_i)\psi(\mathbf{r})d^3\mathbf{r}. \]

Here, the potential \( U \) accounts for the FmnC charge and for band mismatch between normal and magnetic metals, whereas \( J \) is the exchange interaction; \( \mu s \) is the magnetic moment of each cluster; \( \sigma \) stands for the electron spin operator. The MR analysis in materials with large spin clusters, \( s \gg 1 \) can be done using a static exchange field model \( \mathbb{F} \), where the operator \( \mathbf{\hat{s}} \) is replaced by \( s\mathbf{I} \) (here, \( \mathbf{I} \) is a unit vector in the direction of polarisation of an individual cluster). In contrast, the analysis of a thermally excited spin current and the magneto-thermopowers needs to take into account the quantum nature of the cluster spin, manifested through the electron spin-flip process.

The formation of electron-hole asymmetry in the quasiparticle lifetime becomes apparent after analyzing the imaginary part of the self-energy to the lowest non-vanishing order of perturbation theory. Fig. \( \mathbb{F} \) shows the Keldysh diagrams which appear in the self-energy \( \Sigma^A(\omega) \) in the second order of the electron-cluster interaction, after averaging over random positions, \( \mathbf{r}_i \), of the clusters. We assume a thermal distribution of clusters. Together, these diagrams yield the scalar and exchange potential scattering amplitudes (from the same impurity) is taken into account by the diagrams in Fig. 1(c), which is responsible for the spin-dependence of the electron mean free path in the presence of polarised clusters. Together, these diagrams yield

\[ \text{Im} \Sigma^A(\mathbf{\epsilon}) = \pi\gamma n_c \left[ U^2 + J^2 s(s + 1) + 2UJ\langle s_z \rangle \mathbf{\hat{s}}_z \right] - \pi\gamma n_c J^2\langle s_z \rangle \mathbf{\hat{s}}_z \tanh \left( \frac{\mathbf{\epsilon} + \mu B\mathbf{\hat{s}}_z}{2T} \right), \tag{5} \]

where \( n_c \) is the concentration of FmnC’s. Diagrams \( \mathbb{A} \)–(c) describe elastic processes. The spin-flip diagrams \( \mathbb{D} \) contain an inelastic part resulting in the energy-dependent contribution towards \( \text{Im} \Sigma^A \) in Eq. (5), and they are responsible for the kinetic electron-hole asymmetry. This asymmetry is most pronounced when the ensemble of clusters is fully polarised, \( \langle l_z \rangle \rightarrow 1 \), and vanishes when \( \langle l_z \rangle = 0 \).

To explain the origin of this asymmetry and its polarisation dependence we consider the limit of \( \langle l_z \rangle \rightarrow 1 \), so that all FmnC’s are polarised ‘up’. Fig. 2(a) shows that an incident spin-down electron with energy \( \epsilon > \mu B \) above the Fermi level is able to flip its spin turning the spin of FmnC from the polarisation axis. The amplitude of such a process is \( A \sim J\langle s - 1|\mathbf{\hat{s}}_z|s \rangle \sim J\sqrt{s} \), which results in the scattering rate \( \tau^{-1}_s \propto J^2 s \). The relevant range of a quasiparticle excitation energy for this process is set by the FmnC energy splitting, \( \mu B \), between its initial and scattered spin state, which differ by \( s_z \rightarrow -s_z \). A similar process is possible for the incident spin-up hole with \( \epsilon < -\mu B \) below the Fermi level, Fig. 2(d): it corresponds to the reverse process of an equilibrium spin-down electron relaxing into an empty state with opposite spin below the
use the kinetic equation approach. That is, we study a

spin channel results in the generation of a spin current

is inverted in the opposite spin channels, in accordance

resulting behaviour of the quasi-particle lifetime for elec-

A

Fermi level (hole). In contrast, neither spin-up electrons

above the Fermi level, nor spin-down holes shown in Figs.

2(b,c) can scatter with changing spin state, since they

cannot increase further the maximal \( s_z = s \) of a cluster.

All together, these processes determine the energy-

dependent part in the electron scattering rate from fully

polarized clusters,

\[
\text{Im } \Im A \rightarrow \rho^{-1} = \begin{pmatrix}
\frac{1}{2} \tau_{1}^{-1} + \theta(-\varepsilon - \mu B)\tau_{2}^{-1} & 0 \\
0 & \frac{1}{2} \tau_{1}^{-1} + \theta(\varepsilon - \mu B)\tau_{2}^{-1}
\end{pmatrix},
\]

\[
\tau_{1}^{-1} = \frac{2\pi n_{c}}{h} \frac{\gamma}{\varepsilon_{F}^{2}} (U^{2} + J^{2} s(s+1) \pm 2UJ s),
\]

\[
\tau_{s}^{-1} = \frac{4\pi n_{c} c}{h} J^{2}s.
\]

Note that for a cluster with \( N \) magnetic atoms \( S \propto N \)

and \( U \propto N \) so that \( \tau_{1}^{-1} \propto N^{2} \), whereas \( \tau_{s}^{-1} \propto N \). The resulting behaviour of the quasi-particle lifetime for electrons in the presence of polarized FnnC’s is sketched in Fig. 2(e). It indicates that the electron-hole asymmetry is inverted in the opposite spin channels, in accordance

with the symmetry of the Kondo problem 14.

The inverted electron-hole asymmetry in the opposite

spin channel results in the generation of a spin current

when two parts of the system are held at different tempera-

tures. To describe a thermally generated spin-current,

\( j_s = \beta \nabla T \), as well as the electric current \( j = \kappa \nabla T \), we

use the kinetic equation approach. That is, we study a

steady-state kinetic equation,

\[
\frac{\nabla \cdot \kappa E - e \rho \cdot \partial_{\rho}}{\tau_{s}} = \langle \hat{J}(z, s, T) \rangle,
\]

where the collision term \( I_{\pm}(\hat{\rho}, T) \) describes the balance

in the distribution functions \( \rho_{\pm} \) of spin-up and spin-down electrons which scatter from a group of FnnC’s in a given initial spin state, \( s_z \). The brackets \( \langle \ldots \rangle \) stand for averaging over the thermal distribution of cluster spins. The averaged collision term therefore takes into account all
electron scattering processes described by the diagrams in Fig. 1.

\[
I_{\pm} = \frac{2\pi n_{c}}{h} \frac{U \pm J s_z^{2}}{\varepsilon_{F}^{2}} \int \frac{d^{3}\rho'}{h^{3}} \left( \delta_{\rho} - \delta_{\rho_{\pm}} \right) \delta (\varepsilon_{\rho} - \varepsilon_{\rho'})
\]

\[
+ \frac{2\pi n_{c}}{h} \int \frac{d^{3}\rho'}{h^{3}} \left( (s_{z}^{2} - s_{z}^{2} + s_{z} \pm s_{z}) \rho' \left( 1 - \rho_{\pm} \right) - \left( s_{z}^{2} - s_{z}^{2} + s_{z} \pm s_{z} \right) \rho_{\pm} \left( 1 - \rho' \right) \right) \delta (\varepsilon_{\rho'} - \varepsilon_{\rho \mp} \mp B).
\]

The first term in \( I_{\pm} \) describes the elastic spin-conserving processes in Fig. (1a-c), whereas the second takes care of spin-flip processes corresponding to diagrams in Fig. (1d). By inspection, one can see that the probabilities of electrons \( \downarrow \rightarrow \uparrow \) and \( \uparrow \rightarrow \downarrow \) spin-flip scattering from the same cluster with a given \( s_z \) slight differ. In a system where FnnC’s are partially polarised (along the external magnetic field \( B = n_{z} B \)) this leads to the energy- and spin-dependent electron momentum relaxation rate, \( 2 \text{Im } \Im A / \rho^{1} / h \) where \( \alpha = \uparrow, \downarrow \) given by Eq. 6. In the leading order in a small temperature gradient \( \left( \nu_{F} \tau T^{-1} \nabla T \ll 1 \right) \), the spin-current, \( j_{s} = \frac{4}{3} \int \gamma d^{3} \partial \langle \hat{J}_{s} \rangle \) and electric current, \( j = \frac{2}{3} \int \gamma d^{3} \partial \langle \hat{J} \rangle \) are determined by the first angular harmonic in momentum space, \( \hat{J} \) of the electron-density-matrix, \( \hat{J} \approx \hat{J}_{s} + \hat{J}_{s} p / p \). This first angular harmonic can be found using the equation

\[
\frac{2}{h} \text{Im } \Im A \hat{J} = \frac{\nu_{F} \tau T^{-1} \nabla T}{T} \nabla T,
\]

where \( \hat{J}_{s}(\varepsilon) = \left[ e^{is / kT} + 1 \right]^{-1} \) is the Fermi function.

As a result, we calculate the spin-current, \( j_{s} = \beta \nabla T \), which, for large spin clusters, \( s \gg 1 \) is described by

\[
\beta = -\frac{2h c \gamma k^{2}}{\varepsilon_{F}^{2}} \frac{\tau_{s}^{2}}{\tau_{s}} \int \langle l_{z} \rangle x \times \frac{\tau_{s}^{2}}{\tau_{s}} \times \left( \tau_{1} + \tau_{s} \right)^{2} \left( \tau_{1} + \tau_{s} \right)^{2} \left( l_{z} \right)^{2} \left( l_{z} \right)^{2} \left( l_{z} \right)^{2}.
\]

In the above result, scattering rates \( \tau_{1}^{-1} \) and \( \tau_{s}^{-1} \) are defined in Eq. 6, and the factor \( f = \frac{x^{2} e^{-x / c^{2}}}{\left(e^{x / c^{2}} - 1\right)} \) with \( x = \mu B / kT \) is specified for the case \( s \gg 1 \) and takes into account the suppression of the spin transfer rate at low temperatures and high magnetic field, such that \( \mu B > kT \), since it requires the electron energy transfer \( \varepsilon - \varepsilon' = \mu B \). Also, in most of the metals with embedded ferromagnetic clusters, the maximum MR effect
is $\Delta \lesssim 10\%$, which is additionally suppressed by non-magnetic impurities and phonon scattering. Therefore, we can simplify the result in Eq. 8 further, using the fact that $(\tau_\uparrow - \tau_\downarrow) / (\tau_\uparrow + \tau_\downarrow) \ll 1$, which leads to the approximate form in Eq. 11.

Due to the difference, $(\tau_\uparrow - \tau_\downarrow) (l_z)$ between the mean free path times of spin-$\uparrow$ and $\downarrow$ carriers scattering from the ensemble of partially polarised FmnC’s, the spin-current ‘drags’ a charge current $j \propto \nabla \mathcal{N}$. For $s \gg 1$, we find

$$\kappa = \frac{8e^2 v_F^2 \gamma}{3 \tau_s} \left[ \frac{\tau_\uparrow^2 \tau_\downarrow (\tau_\uparrow - \tau_\downarrow)}{\tau_\uparrow + \tau_\downarrow} \right]^2 f \left( \frac{\mu B}{kT} \right).$$

Together with the MR of this material obtained using the same approximations,

$$R = \frac{3}{e^2 v_F^2 \gamma} \frac{\tau_\uparrow + \tau_\downarrow}{4 \tau_\uparrow \tau_\downarrow} \left[ 1 - \frac{\tau_\downarrow - \tau_\uparrow}{\tau_\uparrow + \tau_\downarrow} (l_z)^2 \right],$$

this determines the thermopower coefficient $c = -\kappa R$ with a magnetic field dependence. The magneto-thermopower $\Xi(B) \equiv c(B) - c(0)$ is related to the observable MR as

$$\Xi(B) = \frac{k}{e} \frac{\tau_\uparrow \tau_\downarrow}{\tau_\uparrow + \tau_\downarrow} \frac{\Delta(B)}{1 + \Delta(B)} f \left( \frac{\mu B}{kT} \right).$$

(9)

The magnetic field dependence of $\Xi(B)$, implicit in the above result, contains two field scales $\Xi(B)$. At a low magnetic field where the polarisation of clusters develops, the MTP is proportional to the MR and saturates together with $\Delta(B)$ at the field $B_{MR}$. At a higher field range, $B > kT/\mu$ the MTP is suppressed and dies away. For weak MR materials, $\Delta \lesssim 0.1$, where $\tau_\downarrow - \tau_\uparrow \ll \tau = \tau_\uparrow + \tau_\downarrow$, the above relation can be approximated using the formula in Eq. 9.

At the early stage of study of Kondo impurity in metals, a strong magneto-thermopower has been noticed in various dilute magnetic alloys [13, 14, 15, 16, 17, 18, 19]. Later, a strong magnetic field dependent thermopower has been observed in metals with embedded ferromagnetic nanoclusters [20, 21, 22, 23, 24], where it has been attributed to the energy dependent density of states in a weakly ferromagnetic metal with a complex band structure [3]. However, when viewed in terms of the above-presented theory, these observations indicate the presence of thermally excited spin-current, Eqs. 11-13 generated by a temperature gradient.

Recently, a giant MTP has been measured in copper with embedded Co clusters of a controlled size. It was noticed that MTP weakens with increase of the number of atoms, $N$, in a FmnC [22]. This behaviour as well as the observed direct correspondence between MTP and MR can be explained on the basis of the result in Eq. 11. The matter is that for a cluster with $N$ magnetic atoms $\tau_\uparrow^{-1} \propto N^2$, whereas $\tau_s^{-1} \propto N$. Therefore, for two samples with the same densities of the embedded clusters containing $N$ and $N'$ atoms, we estimate $\Xi(N)/\Xi(N') \propto N'/N$, the MTP is weaker in the system with larger clusters.

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[1] I. Žutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. 76, 323-410 (2004); G.A. Prinz, Science 282, 1660 (1998) and refs. therein
[2] T. Valet and A. Fert, Phys. Rev. B 48, 7099 (1993); D.H. Mosca et al, Journ. of Magn. and Magn. Mat. 94, L1 (1991)
[3] S. Zhang, Appl. Phys. Lett. 61, 1855 (1992); S. Zhang and P.M. Levy, J. Appl. Phys. 73, 5315 (1993); H. Cama-long, S. Zhang, and P.M. Levy, J. Appl. Phys. 75, 6906 (1994), P. Holody et al, Phys. Rev. B 50, 12990 (1994)
[4] A. Berkowitz et al, Phys. Rev. Lett. 68, 3745 (1992); J. Xiao et al, Phys. Rev. B 46, 9266 (1992)
[5] K. Yoshida, Phys. Rev. 107, 396 (1957); H. Rohrer, Phys. Rev. 174, 583 (1968)
[6] I. Korenblit, J. Phys. F 12, 1259 (1982)
[7] L. Piraux et al, Journ. of Magn. and Magn. Mat. 110, L247 (1992); J.L. Duval et al, J. Appl. Phys. 75, 7070 (1994)
[8] E. McCann and V.I. Fal’ko, Appl. Phys. Lett. 81, 3609 (2002); E. McCann and V.I. Fal’ko, Phys. Rev. B 66, 134424 (2002)
[9] L. Xing et al, Phys. Rev. B 48, 6728 (1993)
[10] B. Giovannini and S. Koide, Progr. Theor. Phys. 34, 705 (1965)
[11] J. Kondo, Progr. Theor. Phys. 34, 372 (1965); L.Gurevich and I. Yassievich, Sov. Phys. JETP 20, 922 (1965); K.Maki, Progr. Theor. Phys. 41, 586 (1969); R.Weiner and M. Beal-Monod, Phys. Rev. B 2, 2675 (1970); N. Kawakami, T. Usuki, and A. Okiji, J. Phys. Soc Jpn 56, 1539 (1987)
[12] M.G. Vavilov, L.I. Glazman, and A.I. Larkin, Phys. Rev. B 68, 075119 (2003)
[13] M. Peskin, D. Schroeder, Introduction to Quantum Field Theory, HarperCollins 1995
[14] A. Hewson, The Kondo Problem to Heavy Fermions, Cambridge University Press, 1993
[15] For a metal with spin-$\frac{1}{2}$ impurities, their polarisation and MR develop over the same range of a magnetic field as the field suppressing the electron spin-flip scattering. Therefore, in a metal with spin-$\frac{1}{2}$ Anderson impurities, magneto-thermopower would display a sharp non-monotonic field dependence with a maximum effect at $B \approx 2kT/\mu$, $\Xi = \frac{\tau_\uparrow}{e \tau_\uparrow (\tau_\uparrow - \tau_\downarrow) \cosh^{2}(\mu B/2kT)}$.  
[16] D. Huntley and C. Walker, Can. J. Phys. 47, 865 (1969)
[17] R. Berman et al, Physics Letters A 27, 464 (1968); R. Berman and J. Kopp, J. Phys. F 1, 457(1971); J. Kopp, ibid. 6, 1211 (1975)
[18] M. Read and A. Guenault, J. Phys. F 4, 94 (1974)
[19] L. Azevedo et al, Phys. Rev. B 20, 4450 (1979)
[20] J. Shi, E. Kita, L. Xing, and M. Salamon, Phys. Rev. B 48, 16119 (1993); J. Shi et al, Phys. Rev. B 54, 15273 (1996)
[21] L. Piraux et al, Phys. Rev. B 48, 638 (1993)
[22] H. Sato et al, Journ. of Magn. and Magn. Mat. 152, 109 (1996); H.Sato, Mat. Sci. Eng. B 31, 101 (1995); H.Sato et al, J. Phys. Condens. Matter 7, 7053 (1995)
[23] Y. Kobayashi et al, J. Phys. Cond. Matter 8, 11105 (1996)
[24] J. Sakurai et al, J. Phys. Soc. Jpn. 66, 2240 (1997)
[25] S. Serrano-Guisan et al, Nature Materials AOP, doi:10.1038/nmat1713 (2006)