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Physica E 18 (2003) 64–68
PHYSICA E
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Electron energy and phase relaxation on magnetic impurities

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Abstract

We discuss the effect of magnetic impurities on the inelastic scattering and dephasing of electrons. Magnetic impurities mediate the energy exchange between electrons. This mechanism is especially effective at small energy transfers E in the absence of Zeeman splitting, when the two-particle collision integral in the electron kinetic equation has a kernel $K \propto 1/E^2$ in a broad energy range. In a magnetic field, this mechanism is suppressed at E below the Zeeman energy. Simultaneously, the Zeeman splitting of the impurity spin states reduces the electron dephasing rate, thus enhancing the effect of electron interference on conduction. We find the weak localization correction to the conductivity and the magnitude of the conductance fluctuations in the presence of magnetic field of arbitrary strength. Our results can be compared quantitatively with the experiments on energy relaxation in short metallic wires and on Aharonov–Bohm conductance oscillations in wire rings.

Keywords: Kondo effect; Electron energy relaxation; Weak localization; Conductance fluctuations

1. Introduction

The effect of magnetic impurities on the electron properties of a metal is drastically different from that of "usual" defects which just violate the translational invariance of the crystalline lattice. The reason for the difference is that a magnetic impurity brings an additional degree of freedom—its spin. We demonstrate that magnetic impurities may mediate energy transfer between electrons. If the transferred energy E exceeds the Kondo temperature T_K, then the energy relaxation occurs predominantly in two-electron collisions. We derive the kernel K of the collision integral in the kinetic equation for the distribution function. This kernel depends strongly on the transferred energy, $K \propto J^4/E^2$. The dependence of K on the energies ε_i of the colliding electrons comes from the logarithmic in $|\varepsilon_i|$ renormalization of the exchange integral J, known from the theory of Kondo effect [1], and is relatively weak as long as $|\varepsilon_i| \gg T_K$. The $1/E^2$ divergence of the kernel is cut off at small E; the cut-off energy is determined by the dynamics of the impurity spins.

Localized spins affect not only the energy relaxation rate, but also the conventional electron transport properties, such as the temperature and field dependence of the conductance. No spin dynamics of impurities is needed for the suppression of the interference corrections to the conductivity; interaction of electron spins with the magnetic moments "frozen" in random directions already leads to that suppression [2]. Universal conductance fluctuations (UCF) are not suppressed by "frozen" magnetic moments. However, even a relatively slow relaxation (such as Korringa relaxation) of individual magnetic moments leads to the time-averaging of the random potential "seen" by transport electrons in the course of measurement, and the mesoscopic fluctuations of the DC conductance are averaged out [3].[1] We find the weak localization correction to the conductivity and the magnitude of conductance fluctuations in the presence of magnetic field of arbitrary strength.

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[1] The limits of zero and strong $\omega_s \gg T$ are well studied, see Ref. [3].

1386-9477/03/$ - see front matter
doi:10.1016/S1386-9477(02)00974-8

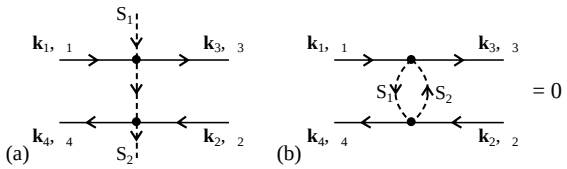

Fig. 1. (a) A characteristic diagram for the amplitude of inelastic electron–electron scattering mediated by the exchange interaction of electrons with a magnetic impurity, in notation of Ref. [6]. The solid lines denote electron states, the dashed lines denote the localized spin state. (b) The scattering amplitude cannot be represented in the form of an effective four-electron vortex.

2. Inelastic scattering of an electron off a magnetic impurity

We describe a metal with magnetic impurities by means of the Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V}$:

$$\hat{H}_0 = \sum_{\mathbf{k}\alpha} \xi_{\mathbf{k}} c^\dagger_{\mathbf{k}\alpha} c_{\mathbf{k}\alpha}, \quad \hat{V} = J \sum_{\alpha\alpha',l} \hat{\mathbf{S}}_l \boldsymbol{\sigma}_{\alpha\alpha'} \psi^\dagger_{\mathbf{r}_l\alpha} \psi_{\mathbf{r}_l\alpha'}, \tag{1}$$

where $\hat{\mathbf{S}}_l$ is the spin operator of the lth impurity at point $\mathbf{r}_l$, $\hat{\mathbf{S}}_l^2 = S(S+1)$. Free electron states $c_{\mathbf{k}\alpha}$ are labelled by the wave vector $\mathbf{k}$ and the spin index α, $\psi_{\mathbf{r}_l\alpha} = \sum_{\mathbf{k}} e^{i\mathbf{k}\mathbf{r}_l} c_{\mathbf{k}\alpha}$. The Pauli matrices are denoted by $\boldsymbol{\sigma} \equiv (\sigma^x, \sigma^y, \sigma^z)$.

The impurities can be considered independently if their concentration n is low enough. In the one-impurity scattering problem, there is interaction only in s channel, so we will label the participating electron states with scalar index k.

In the framework of the exchange Hamiltonian (1), the lowest non-vanishing order of the perturbation theory series in the exchange constant J for the inelastic scattering amplitude is the second order:

$$A^{SS'}_{\varrho_1\varrho_2;\varrho_3\varrho_4} = \langle \varrho_3\varrho_4, S' | \hat{V} \frac{1}{\xi_{k_1} + \xi_{k_2} - \hat{H}_0} \hat{V} | \varrho_1\varrho_2, S \rangle, \tag{2}$$

where $\varrho_i = (k_i, \alpha_i)$. In the diagrammatic representation, the amplitude is the sum of the diagram shown in Fig. 1a and the other three diagrams that can be obtained from the diagram of Fig. 1a by the transposition of indices $1 \leftrightarrow 2$ and/or $3 \leftrightarrow 4$. Note that there is no summation over the initial or final spin states of the impurity in Eq. (2). Thus the spin lines are not closed, i.e. contrary to Ref. [4] this scattering amplitude cannot be represented in the form of an effective four-electron vortex (Fig. 1b).

The denominator in Eq. (2) is the energy of the intermediate virtual state, which equals $\pm(\xi_{k_1} - \xi_{k_3})$ for two of the four possible pairings of the electron creation-annihilation operators (one of these pairings is shown on Fig. 1a), or $\pm(\xi_{k_1} - \xi_{k_4})$ for the other two pairings. The spin structure of the scattering amplitude can easily be found from Eq. (2). In a scattering event, spins of one or both participating electrons must flip, with the corresponding change of the impurity spin. Here we are interested only in the relaxation of the electron energy distribution, and assume that in the absence of magnetic field the system does not have any spin polarization. Therefore we need to calculate only the total cross-section of scattering into all possible spin states, averaged over the initial spin states of the impurity and two electrons. We obtain the collision integral kernel

$$K(E) = \frac{\pi}{2} \frac{n}{\nu} S(S+1)(J\nu)^4 \frac{1}{E^2} \tag{3}$$

which depends only on the energy E transferred in the collision. Here ν is the electron density of states at the Fermi energy per spin degree of freedom.

For low energy electrons, the effective exchange constant J is renormalized due to the Kondo effect [5]. In the leading logarithmic approximation [6] the renormalized expression for the exchange constant in Eq. (3) is

$$J = \frac{2}{\nu} \ln^{-1} \frac{\varepsilon^*}{T_K}, \tag{4}$$

where ε^* is the characteristic energy of electrons participating in the collision and T_K is the Kondo temperature. This approximation is justified as long as the energies $\varepsilon_i \sim \varepsilon^*$ of all incoming and outgoing electrons satisfy the condition $\varepsilon^* \gtrsim T_K$. It is important to note that energy ε^*, which lies within the width of the electron distribution function, does not cut off the singularity in the transferred energy E. For a more detailed expression for the renormalized $K(E)$ see Ref. [7].

The low-energy divergence of the inelastic scattering amplitude (2) is cut off by the time evolution of the impurity spin correlator $\langle S' | \hat{S}^j(t) \hat{S}^k(t') | S \rangle$. In magnetic field B this evolution is a spin precession with frequency $\omega_s = g_{\text{imp}} \mu_B B$. When ω_s exceeds the energies of the electrons being scattered, the scattering rate saturates [8] at

$$K(E) \sim \frac{n}{\nu} S(S+1)(J\nu)^4 \frac{1}{\omega_s^2}. \tag{5}$$

The scattering processes in which both initial or both final electrons have the same spin are suppressed completely.

The other mechanism, which cuts off the $E = 0$ singularity of the kernel (3) even at $B = 0$, is the impurity spin relaxation. The relaxation limits the lifetime of the intermediate state and the denominator in Eq. (2) acquires the imaginary part. At high temperature $T > T_K$ scattering of the thermal electrons on the spin results in an exponential decay of the spin correlation function, $\langle S' | \hat{S}^j(t) \hat{S}^k(t') | S \rangle \propto \exp(-|t - t'|/\tau_T)$. The impurity spin correlation time τ_T can be evaluated with the help of the Fermi golden rule. If the deviation from the thermal equilibrium is weak, we have

$$\frac{\hbar}{\tau_T} = \frac{2\pi}{3} (J\nu)^2 T\ . \tag{6}$$

As T is lowered towards T_K, the exchange constant is renormalized according to Eq. (4). The resulting expression for

the spin-flip rate reads:

$$\frac{\hbar}{\tau_T} = \frac{2\pi}{3}[\ln(T/T_K)]^{-2}T. \tag{7}$$

The energy scale $\hbar/\tau_T$ sets the limit of applicability of Eq. (2) and cuts off the singularity in the kernel (3) at $E \sim \hbar/\tau_T$. Note that within the limits of applicability of Eq. (7), the spin-flip rate satisfies the condition $T > \hbar/\tau_T > T_K$.

At very small energies ($|\varepsilon_i|, T \ll T_K$) the Fermi-liquid description of electrons is valid again. The behavior of the system is described in this case by the quadratic fixed-point Hamiltonian, with the four-fermion interaction being the least-irrelevant term [9,10]. The calculation of the inelastic scattering rate is then straightforward, the resulting collision-integral kernel is given by

$$K(E) = \frac{1}{T_K^2}\frac{n}{v}. \tag{8}$$

When $T = 0$, the inelastic scattering rate is $\hbar/\tau_{in} = \int_0^\varepsilon dEK(E)E \propto (\varepsilon/T_K)^2$ and decreases faster than ε at small $\varepsilon \to 0$, as it is supposed to be in the Fermi-liquid picture.

We also discuss the relaxation due to the electron scattering on magnetic impurities in wires with applied bias $eV \gg T$. In this case the electron distribution is smeared, and the width of smearing eV exceeds the typical energies $|\varepsilon_i|$ of the colliding electrons. Assuming $eV \gg T_K$ and substituting the renormalized exchange constant $J = 2/(v\ln(eV/T_K))$ into the kernel Eq. (3), we obtain

$$K(E) = \frac{\pi}{2}\frac{n}{v}S(S+1)[\ln(eV/T_K)]^{-4}\frac{1}{E^2}. \tag{9}$$

The $1/E^2$ dependence in Eq. (9) persists down to the cut-off, which is determined by the spin-flip rate $1/\tau_{eV}$. For the spin-flip rate in the non-equilibrium situation the temperature T in Eq. (7) should be replaced by the electron distribution function smearing eV:

$$\frac{\hbar}{\tau_{eV}} = \gamma[\ln(eV/T_K)]^{-2}eV. \tag{10}$$

Here the numerical constant $\gamma \sim 1$ depends on details of the non-equilibrium electron distribution function.

3. Effect of spin scattering on electron interference phenomena

Magnetic impurities provide mechanism not only for electron energy relaxation but also for electron phase relaxation, which suppresses the interference phenomena, such as weak localization and conductance fluctuations. Here we consider these phenomena for metal wires with magnetic impurities, which can be partially polarized by an applied magnetic field [3].

The weak localization correction to the conductivity of a wire without spin-orbit scattering in the conditions of strong Zeeman splitting of the conduction electron states ($\varepsilon_Z\tau_s \gg 1$) and slow impurity spin relaxation ($\tau_T \gg \tau_s$) is [11]

$$\Delta\sigma = -\frac{e^2}{4\pi\hbar T}\int \frac{d\varepsilon}{\cosh^2\varepsilon/2T}\frac{\sqrt{D\tau_s}}{\sqrt{P(\varepsilon)+B^2/B_c^2}}. \tag{11}$$

Here $1/\tau_s = 2\pi vnJ^2S(S+1)$ is the scattering rate of electrons on magnetic impurities in the absence of magnetic field. Function $P(\varepsilon)$ represents the probability of spin flip in the presence of magnetic field B:

$$P(\varepsilon) = 1 - \frac{\langle\hat{S}_z^2\rangle + \langle\hat{S}_z\rangle\tanh(\varepsilon+\omega_S)/2T}{S(S+1)} \tag{12}$$

For $S = \frac{1}{2}$ impurities, we have $\langle\hat{S}_z^2\rangle = 1/4$ and $\langle\hat{S}_z\rangle = (1/2)\tanh(\omega_s/2T)$. In this case function $P(\varepsilon)$ can be rewritten in the form:

$$P(\varepsilon) = \tfrac{4}{3}(p_\downarrow(1-n(\varepsilon+\omega_S)) + p_\uparrow n(\varepsilon+\omega_S)), \tag{13}$$

where $p_{\uparrow,\downarrow} = e^{\pm\omega_S/2T}/(2\cosh\omega_S/2T)$ is the probability for the impurity spin to be parallel (antiparallel) to the direction of the magnetic field and $n(\varepsilon) = (1+\exp(\varepsilon/T))^{-1}$ is the electron occupation number with energy ε.

The term B^2/B_c^2 in Eq. (11) represents the orbital effect of the applied magnetic field on conducting electrons; B_c defines the characteristic value of the magnetic field, which produces the orbital dephasing rate comparable with the spin scattering rate:

$$B_c = \vartheta\frac{\Phi_0}{\sqrt{D\tau_s A_w}}, \qquad \Phi_0 = \frac{2\pi\hbar c}{e}. \tag{14}$$

Here A_w is the wire cross-section area and ϑ is a dimensionless factor depending on the wire geometry and the magnetic field orientation. The expression in the denominator $\sqrt{D\tau_s A_w}$ represents the effective area, covered by an electron trajectory between consequent spin flips. The characteristic magnetic field B_c gives an upper estimate on system temperature T_c, below which the effect of spin polarization prevails the orbital effect of magnetic field:

$$T_c = Sg_{imp}\mu_B B_c. \tag{15}$$

If the orbital effect of the magnetic field is strong, we expand Eq. (11) in B_c/B and obtain:

$$\Delta\sigma = -\frac{e^2}{\pi\hbar}\sqrt{D\tau_s}\left(\frac{B_c}{B} - \frac{2}{3}\frac{B_c^3}{B^3}\frac{\omega_S}{T}\sinh^{-1}\frac{\omega_S}{T}\right). \tag{16}$$

Conductance fluctuations can be considered similarly to the evaluation of $\Delta\sigma$. We concentrate here on the amplitude of the Aharonov–Bohm "hc/e" oscillations. Magnetic field applied through the ring changes electron wave functions and, consequently, the conductance of the ring of radius r. The conductance statistics is characterized by the correlation

function:

$$\langle\langle g_\Phi g_{\Phi+\Delta\Phi}\rangle\rangle = \alpha \frac{e^4}{\hbar^2} \sum_{k=0}^{\infty} R_k \cos 2\pi k \frac{\Delta\Phi}{\Phi_0}, \tag{17}$$

where $\Phi = \pi r^2 B$ is the magnetic flux through the ring and α is a dimensionless geometry dependent factor.

In the high temperature case,[2] $\tau_s T \gg 1$, we find the amplitude of oscillations of the conductance correlation function [11]:

$$R_k = \frac{D^{3/2}}{r^3 T^2} \int \frac{e^{-2\pi k r\sqrt{\Gamma(\varepsilon)/D}}}{\sqrt{\Gamma(\varepsilon)}} \frac{d\varepsilon}{\cosh^4 \varepsilon/2T},$$

$$\Gamma(\varepsilon) = \gamma + \frac{1}{\tau_s} \times \left(1 - \frac{\langle\hat{S}_z\rangle^2 + \langle\hat{S}_z\rangle \tanh(\varepsilon + \omega_S)/2T}{S(S+1)}\right) \tag{18}$$

with γ being the dephasing rate due to mechanisms other than magnetic impurity scattering.

4. Comparison with experiments

Relaxation of the electron energy distribution was investigated experimentally in metallic wires of Cu and Au in Refs. [12,13]. In these experiments, a finite bias V was applied to the wire terminals. It was found that starting from fairly small wire lengths, the electron distribution is smeared over the range of energies eV, instead of having two distinct steps created by the bias applied to the wire ends. The observed electron energy relaxation was attributed [12,13] to electron–electron collisions. The collision-integral kernel for $E < eV$ extracted from the experiments has the form $K(E) = \hbar/(\tau_0 E^2)$, with a cut-off at some low energy, which scales linearly with eV [14].

Properties of these samples are compatible with the presence of iron impurities with a concentration up to few tens of ppm [14]. The spin-flip rate, Eq. (10), is the low-energy cut-off for the $1/E^2$ kernel dependence. This cut-off is roughly proportional to the applied voltage, in agreement with experimental observations [14]. We must note, however, that the lower voltages used in experiment [13] are close to the Kondo temperature, so the leading-logarithmic approximation [6,15], used in derivation of Eqs. (9) and (10), may be insufficient.

Recent experiments [16] demonstrate that the previously observed [12] electron energy relaxation in thin wires is indeed suppressed by the applied magnetic field, thus supporting our hypothesis that the origin of the relaxation is the inelastic scattering on the magnetic impurities.

[2] We notice that even if the spin scattering is strong initially, $\tau_s T \ll 1$, due to the impurity spin polarization, the effective scattering rate decreases at sufficiently strong magnetic field, leading the system to the high temperature regime.

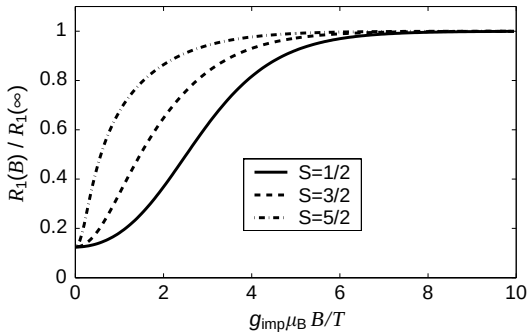

Fig. 2. Figure shows dependence of the "hc/e" oscillations of the conductance correlation function on the applied magnetic field B for several values of the impurity spin S in case when $\gamma\tau_s = 1.5$.

In measurements [17] of the conductance of a Cu ring, the amplitude of the conductance oscillations increases in strong magnetic field $\omega_c \sim T$. This observation can be explained as the result of the impurity spin polarization by the magnetic field. Fig. 2 represents the amplitude of the first harmonic ("hc/e" oscillations) of the conductance correlation function in the limit $T \gg \gamma,\ 1/\tau_s$, described by Eq. (18), for different values of the impurity spin S.

In conclusion, the exchange interaction of itinerant electrons with magnetic impurities can facilitate electron energy and phase relaxation. We derived the kernel of the collision integral which determines the energy relaxation, and found the weak localization correction to the conductivity and the amplitude of conductance fluctuations at an arbitrary level of polarization of magnetic impurities by an external magnetic field. The obtained results provide a quantitative explanation of the experiments [12,13] on anomalously strong energy relaxation in short metallic wires and may be compared with the observed behavior of the "hc/e" oscillations of the conductance of an Aharonov–Bohm ring [17].

Acknowledgements

The authors are grateful to I. Aleiner, N. Birge, and H. Pothier for valuable discussions. This work was supported by NSF Grants No. DMR 97-31756 and DMR 0120702 at the University of Minnesota and by the US-ONR, the LPS, and DARPA at the University of Maryland.

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