Spin-memory effect and negative magnetoresistance in hopping conductivity

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We propose a mechanism for negative isotropic magnetoresistance in the hopping regime. It results from a memory effect encrypted into spin correlations that are not taken into account by the conventional theory of hopping conductivity. The spin correlations are generated by the nonequilibrium electric currents and lead to the decrease of the conductivity. The application of the magnetic field destroys the correlations thus enhancing the conductance. This effect can occur even at magnetic fields as small as a few gauss.

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In strongly disordered conductors, where electronic states are localized, the conduction is due to phonon-assisted tunneling between localized states [1]. The magneto-resistance (MR) in such hopping regime is not well understood. In many insulators the relative magnitude of MR is significantly larger than that of metals [2], and its features are less universal. Experimental measurements in the hopping regime showed both positive (see [1, 4, 11] and references therein) and negative MR [5–12]. In some materials more complicated behavior was observed: a giant MR that changes its sign from positive to negative as the magnetic field increases [1, 12].

The mechanisms that were suggested for hopping MR can be roughly divided into two classes: orbital related and spin related mechanisms. The orbital mechanism is associated with the modification of the hopping amplitude by the magnetic field and, depending on the model, leads to positive [1, 3] or negative [4, 15] MR. The characteristic magnetic field in these cases, \( H^* \), \( A \sim \Phi_0 = \hbar c/e \), corresponds to a flux quanta threading the effective area, \( A \), explored by an electron during the tunneling event. A distinctive feature of the orbital mechanism in two dimensional films is its anisotropy with respect to the direction of the magnetic field.

The spin mechanisms for positive MR are related either to the reduction of the density of states with the increase of the magnetic field due to its effect on doubly occupied states [16], or to a possible reconstruction of the state of the system, see discussion in Refs. [14, 17]. Both mechanisms produce isotropic MR in films, and the characteristic magnetic field, \( \mu_B H^* \sim \min(T, J) \), is obtained from the competition between the magnetic energy of the spin, \( \mu_B H \) (\( \mu_B \) is the Bohr magneton), and either the temperature \( T \) or the exchange energy between spins, \( J \).

The aforementioned theoretical studies predict rather high characteristic magnetic fields, \( H^* \). Also the experiments were mostly focused on relatively high fields.

In this work we propose a spin-related mechanism for negative MR which takes place at weak magnetic fields sometimes as small as one gauss. It is isotropic and emerges from the long memory of nonequilibrium spin correlations created in course of electron transport. Our discussion will be mainly focused on the experimentally relevant regime where the characteristic hopping time, \( \tau \), is much shorter than the spin relaxation time, \( \tau_s \). In this regime the low magnetic field dependence of the conductance is demonstrated in Fig. 1, and is determined by two characteristic fields:

\[
H^{**} = \frac{1}{\delta g \mu_B \tau}, \quad \text{and} \quad H_s = \frac{1}{\delta g \mu_B \tau_s},
\]

where \( \delta g \) is the typical spatial fluctuation in \( g \)-factor. For the hopping conductivity, \( \tau \) exponentially increases as the temperature decreases, therefore, in general \( H^{**} \ll H^* \).

A qualitative explanation of the negative MR due to memory effect is the following: Consider a situation where the hopping rate of an electron between sites \( i \) and \( j \) depends on the relative spin configuration of the hopping electrons and a spin located nearby at site which we denote by \( ij \) (see Fig. 2). In the presence of current
flowing through the system, a nonequilibrium correlation between the spins at sites $i$ and $ij$ is created. For example, an electron approaching site $i$ from the bulk and making an unsuccessful attempt to hop onto site $j$ will diffuse away and its spin density matrix will depend on the spin at site $ij$. If it returns and attempts to hop one more time, this attempt is not purely probabilistic. It is sensitive to the previous history of the system, e.g. if the tunneling electron forms a triplet state with the localized spin then it will still be in a triplet state for the second attempt (for $H = 0$), even though the tunneling between the sites is inelastic. Let us now apply the magnetic field and neglect the spin relaxation for a moment. If all the spins rotated in the same manner, the triplet state would always remain triplet and there would be no magnetoresistance. However, in strongly disordered systems the $g$-factor is random: This implies that the spins at different locations precess in different manners and therefore the spin correlations are destroyed by the magnetic field. The characteristic field where the MR saturates, $H^{**}$, is obtained from the condition that the phase difference between the spins, accumulated on a time scale of the order of the hopping time $\tau$, is of order one. Moreover, the return probability of an electron moving on the Miller-Abrahams network [18] decays algebraically as a function of time. As a result the magnetoconductance exhibits a singular behavior at small magnetic field. The spin relaxation introduces an upper cutoff on the return time, and removes this singularity.

Although our mechanism is of general character [19] (spin-dependent hopping rate is allowed by symmetry), to illustrate the effect we study a simple model where hops from site to site may occasionally also involve a virtual transition through an occupied state, as illustrated in Fig. 2. We shall refer to the spin of the electron on the occupied state as the “link spin” and denote it by $s_{ij}/2$: $s_{ij}^2 = 3$. The probability of passing from $i$ to $j$ has an interference contribution of the direct transition and the indirect transition which takes place when the link spin and the spin of the moving electron form a singlet. We assume that the transition rate associated with interference is small compared to the rate of direct transition, and treat it to leading order in perturbation theory. To further simplify the problem we assume that these link spins are rare and that the concentration of electrons is very low, namely the average occupation of each site (except the link spins) is much smaller than one. We also neglect the effects of the long range Coulomb interactions [1], which are crucial for the temperature dependence of the hopping transport but seem to be less important for our mechanism of MR.

Let $P^0_i$, denote the probability of having no electron on site $i$ while it is occupied by one electron its state is described by a $2 \times 2$ matrix, $\hat{P}^1_i$, in the spin space. These quantities satisfy the normalization condition $P^0_i + tr \hat{P}^1_i = 1$ and therefore one may parametrize the state of an electron at site $i$ by $n_i = Tr \hat{P}^1_i$ and $S_i = Tr \left( \sigma \hat{P}^1_i \right)$, where $\sigma$ are the Pauli matrices.

Average occupation numbers, $\langle n_i \rangle$, are determined by

$$
\frac{d\langle n_i \rangle}{dt} = -\sum_j \left[ \langle n_i \rangle + \gamma_{ij} \left( \langle n_i \rangle - \langle S_i \cdot s_{ij} \rangle \right) \right]\frac{1}{\tau_{i\rightarrow j}} - \langle i \leftrightarrow j \rangle,
$$

(2a)

where $1/\tau_{i\rightarrow j}$ denotes the bare transition rate from site $i$ to site $j$. The first term on the right hand side accounts for the decrease of the average occupation due to hop from site $i$ to site $j$. It contains two contributions. The first $n_i/\tau_{i\rightarrow j}$ is associated with the direct transition, for simplicity we assumed $n_k \ll 1$ and neglected the factor $1-n_k$. The effect of the correlations in $\langle n_i n_j \rangle$ was considered before [20] and it does not change the MR. The second contribution, proportional to $\gamma_{ij} \left( \langle n_i \rangle - \langle S_i \cdot s_{ij} \rangle \right)$, is associated with the interference term of going through the virtual state [we define $s_{ij} \equiv s_{ji}$, $\gamma_{ij} \equiv \gamma_{ji}$]. This transition occur only for the moving electron and the local spin forming a singlet. The corresponding contribution is proportional to the small parameter of indirect transition $|\gamma_{ij}| \ll 1$. The $\langle i \leftrightarrow j \rangle$ term describes the transition from site $j$ to site $i$.

The electron spin dynamics is described by

$$
\frac{d\langle S_i \rangle}{dt} = \mathbf{h}_i \times \langle S_i \rangle - \sum_j \left[ \langle S_i \rangle + \gamma_{ij} \left( \langle S_i \rangle - \langle S_i \cdot s_{ij} \rangle \right) \right]\frac{1}{\tau_{i\rightarrow j}} - \langle i \leftrightarrow j \rangle,
$$

(2b)

with the first term describing the spin precession [$\mathbf{h}_i = \hat{g}_i \mu_B \mathbf{H}$ is local field acting on the electron spin at site $i$, and $\hat{g}_i$ is the corresponding gyromagnetic tensor], and

![FIG. 2: A simplified hopping model in which electron may hop directly between two neighboring sites or go indirectly by forming a virtual singlet state with an electron at a nearby occupied site. The spins of the moving electron, $S_i$, and the localized electron, $s_{ij}$, precess around different local fields, $h_i$ and $h_{ij}$, due to the spatial fluctuations of the $g$-factor.](image-url)
the second line describes the same hopping processes as in Eq. (2c) (we neglect the direct effect of the magnetic field on the hopping rates $Q_{ij}$). Finally, the dynamics of the link spin is a pure precession:

$$\frac{d(s_{ij})}{dt} = h_{ij} \times (s_{ij}),$$

(2c)

where $h_{ij} = g_{ij} \mu_B H$. Exchange fields and the relaxation of the spin via hopping involve terms of the order of $\gamma_{ij}^2$ which we neglect. Other mechanisms of the spin relaxation will be included later through the phenomenological relaxation time, $\tau_s$.

The relation between Eqs. (2) and the corrections to Miller-Abrahams network can be understood as follows: In equilibrium $(S_i \cdot s_{ij}) = 0$, and detailed balance implies $n_i^{eq}/\tau_{i \rightarrow j} = n_j^{eq}/\tau_{j \rightarrow i}$, where $n_i^{eq}$ denotes the equilibrium occupation number. We redefine the variables describing the nonequilibrium state of the system:

$$(n_i) \rightarrow n_i^{eq}(1 + \psi_i), \quad S_i \rightarrow n_i^{eq} S_i, \quad s_{ij} \rightarrow s_{ij},$$

(3)

Equation (2a) reduces to

$$n_i^{eq} \frac{d\psi_i}{dt} = \sum_j \frac{1}{\tau_{ij}} \left[ \psi_j - \psi_i - \gamma_{ij} \left( \langle \hat{S}_j - S_i \rangle \cdot s_{ij} \right) \right],$$

(4)

where we defined

$$\frac{1}{\tau_{ij}} = \frac{n_i^{eq}(1 + \gamma_{ij})}{\tau_{i \rightarrow j}} = \frac{n_j^{eq}(1 + \gamma_{ij})}{\tau_{j \rightarrow i}}$$

(5)

as the equilibrium transition rate between sites $i$ and $j$. If there were no link spins, $\gamma_{ij} = 0$, Eq. (1) would describe the Miller-Abrahams random resistor network with the conductance of the link $i \rightarrow j$ given by $e^2/(2\pi \tau_{ij})$. The essence of the memory effect is that symmetry allows nonequilibrium spin correlations to be a linear function of the occupation numbers, i.e. assuming locality,

$$\gamma_{ij} \left( \langle \hat{S}_j - S_i \rangle \cdot s_{ij} \right) = Q_{ij}(H)(\psi_j - \psi_i),$$

(6)

where $Q_{ij}(H)$ is a function of the magnetic field, $H$. Therefore, as follows from (1) and (3), the conductances are:

$$G_{ij} = \frac{e^2}{\tau_{ij}} \left[ 1 - Q_{ij}(H) \right].$$

(7)

In order to calculate the function $Q_{ij}(H)$, we need the equation for the correlator $C_{\alpha \beta}^{ij} = \langle S_i^\alpha s_{ij}^\beta \rangle$ (where $\alpha, \beta = x, y, z$ label components of the corresponding vectors). The easiest way to obtain the equation is to remove the $\langle . . \rangle$ in Eqs. (2) by multiplying Eq. (2a) by $s_{ij}^\alpha$ and Eq. (2b) by $S_i^\beta$, add the results and average them again. According to Eq. (2c), even in nonequilibrium, $\langle s_{ij}^\alpha s_{ij}^\beta \rangle = \delta_{\alpha \beta} \delta_{ij}$. Then, to the leading order in $\gamma_{ij}$, we obtain [in the variables (3)]

$$n_i^{eq} \left[ \left( \frac{d}{dt} + \frac{1}{\tau_s} \right) C_{\alpha \beta}^{ij} - \epsilon_{\alpha \gamma \delta} h_i^\gamma C_{\gamma \beta}^{ij} - \epsilon_{\beta \gamma \delta} h_j^\gamma C_{\alpha \delta}^{ij} \right] = - \sum_{k \neq l} C_{\alpha \beta}^{ij} - C_{\alpha \beta}^{kl} + \gamma_{ij} \delta_{\alpha \beta} (\delta_{il} - \delta_{jl}) \psi_i - \psi_j,$$

(8)

with $\epsilon_{\alpha \beta \gamma}$ being the antisymmetric tensor, and repeated indices should be summed over. The relaxation time $\tau_s$, introduced phenomenologically, describes all the spin non-conserving processes. The last term is the source of nonequilibrium spin correlations which after generation propagates by diffusion on the Miller-Abrahams network and prescess in the spatially varying local field.

Equations (2a) and (3) form the complete description of the transport for a fixed realization of the relaxation rates and local fields. To obtain the physical conductivity one needs to average over such realizations. It is done using the percolation theory approach to the hopping conductivity $\Pi$ as we describe below.

We notice, that if there were no randomness in the $\hat{g}_i$-tensors the relevant quantity $C_{\alpha \beta}^{ij}$ would not depend on the magnetic field at all, as all spins rotate in the same manner. The correlation function (8) is affected only by the fluctuations of both $\hat{h}_i$ and $h_{ij}$ (i.e. the averaged field may be subtracted). The diffusing spin (index $l$) experiences fluctuating field because it hops from site to site, and, therefore, its accumulated rotation is proportional to the square root of time. On the other hand the field on the link $h_{ij}$ remains stationary and its effect is linear in time. Thus, we can substitute $h_{ij} \rightarrow 0$, $n_i^{eq} h_{ij} \rightarrow n_i^{eq} h_{ij} \bar{z}$, $n_i^{eq}/\tau_s \rightarrow n_i^{eq}/\tau_s$ in Eq. (3) where $\bar{z}$ is the ensemble average of the equilibrium occupation numbers. With this simplification, the function $Q_{ij}$ from Eq. (7) can be related to the properties of the diffusion on the same Miller-Abrahams network. Consider the probability, $P_{mm'}(t)$, to find the particle at the time $t$ on the site $m$ provided that at $t = 0$ it was on $m'$:

$$\frac{dP_{mm'}(t)}{dt} + \sum_n \frac{P_{mm'}(t) - P_{nn'}(t)}{\tau_{mn}} = \delta(t) \delta_{mm'};$$

(9)

Solving Eq. (3) for the stationary case, and substituting the result into Eq. (6), we find

$$Q_{ij}(H) = \langle s_{ij}^\alpha s_{ij}^\beta \rangle = \frac{(\gamma_{ij})^2}{\tau_{ij}} \sum_{l = 1}^\infty \int_0^\infty dt e^{-i \bar{z} (d + h + \tau_s)} \Delta_{ij}(t);$$

$$\Delta_{ij}(t) = \Pi_{ii}(t) + \Pi_{jj}(t) - \Pi_{ij}(t) - \Pi_{ji}(t).$$

(10)

Equation (10) enables us to draw important conclusions about the magnitude of the MR, the characteristic fields and its asymptotic behavior. Indeed, $\Delta_{ij}(t) \approx 2, t \lesssim \tau_{ij}$, and, as we will see later, $t \Delta_{ij}(t) \rightarrow 0$, in the limit $t \rightarrow \infty$. This means that the total magnitude of the integral in Eq. (10) is determined by short
time, and the MR saturates at $\bar{\sigma}(H) \approx 1$ [see Eq. (1)] with $\tau = \gamma_{ij}$, thus $Q_{ij}(H \to \infty) \approx Q_{ij}(0)/3$. As $\gamma_{ij} \ll 1$, the effect on each resistor is small, so that one can always recalculate the change of the observable conductivity in terms of the average change of the conductances of the percolation network:

$$\frac{\sigma(H \to \infty) - \sigma(0)}{\sigma(0)} \sim A = \rho_\gamma^2. \quad (11)$$

where $\rho$ is the probability of having a link spin between two sites on the percolation cluster and the overbar denotes ensemble averaging.

Our calculations relied on the assumption that the number of occupied sites is small, $\rho \ll 1$, and that the amplitude for transition through a virtual state is also small, $|\gamma_{ij}| \ll 1$, which implied that $A \ll 1$. However, in general, these parameters need not be small and both can be of order unity. In this case the magnitude of the memory effect is also of order unity.

The actual value of the saturation magnetic field $H^{**}$ from Eq. (11) strongly depends on the hopping time and may be anomalously small. Consider, e.g., a two-dimensional sample with the resistance $R \sim 10^8 \Omega$. Then, the typical hopping rate is $h/(T \tau) \approx 10^{-6}$ [see Eq. (7)], and $\delta \Gamma \mu B H^*/T \approx 10^{-6}$. Now estimating $\delta \Gamma \sim 0.01$, we obtain $\mu B H^*/T \gtrsim 10^{-4}$ which corresponds to the fields of the order of gauss at $T \approx 1K$.

Let us discuss the MR at $H < H^{**}$. The hopping rates $\tau_{ij}$ are exponentially distributed and the observable conductivity and diffusion are determined by sites belonging to the percolation cluster. Studies of anomalous diffusion on the percolation cluster concluded that

$$\Delta_{ij}(t) = \left( \frac{\tau_{ij}}{\tau} \right)^{1+2d_s/2}, \quad t \gtrsim \tau, \quad (12)$$

where $d_s$ is referred to as a spectral dimension of the percolation cluster (see e.g. Ref. [21] for a review). For spatial dimensions $d = 2, 3$, $d_s$ is close to $1.3$ [21].

Substituting Eq. (12) into Eq. (11), we find

$$\frac{\delta \sigma(H)}{A \sigma(0)} \sim -\Gamma \left( \frac{d_s}{2} \right) \sum_{l=0}^\infty \left[ \left( \frac{\tau_{ij}}{\tau} \right)^{l+2} - \left( \frac{\tau}{\tau_s} \right)^{l+2} \right], \quad (13)$$

where $\Gamma(x)$ is the gamma-function and only the singular dependence for even $d_s$ is retained $[-\Gamma(-0.65) = 3.9]$. The resulting magnetoconductance is sketched in Fig. 1. Strictly speaking, the correlation length of the percolation cluster on the Miller-Abrahams network is infinite only in the limit $T \to 0$. Taking into account the finite correlation radius introduces the new value of the characteristic field below which one has to replace $d_s$ by $d$. It is interesting to point out that for small fluctuation of the $g$-factor, $H^{**} \to \infty$, and Eq. (13) predicts a positive MR via direct dependence of the spin relaxation rate on the magnetic field $\tau_s(H)/\tau_s(0) = 1 + (H/H_*)^2$, where $H_* \sim 1.4$ is determined by the correlation time of a spin relaxation process [22].

To conclude, we considered a minimal model of the negative MR due to memory effects in the hopping regime. Even though within our model, the amplitude of the effect is small, it has the strongest non-analytic magnetic field dependence and the characteristic fields smaller than that for all the other mechanisms considered in the literature. Further interesting development may be in the direction of the more detailed study of the variable range hopping regime where the number of the link spins within the hopping length becomes large. In this case, the memory mechanism is expected to affect not only the preexponential factor of the conductivity but the exponent itself resulting in giant memory MR.

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