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Phys. Rev. B 93, 045206 — Published 19 January 2016

DOI: 10.1103/PhysRevB.93.045206
Influence of spin dynamics of defects on weak localization in paramagnetic 2D metals

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(Dated: December 23, 2015)

Spin-flip scattering of charge carriers in metals with magnetic defects leads to the low-temperature saturation of the decoherence time, [\(\tau_s\)], of electrons at the value comparable to their spin relaxation time, [\(\tau_s\)]. In two-dimensional (2D) conductors such a saturation can be lifted by an in-plane magnetic field, [\(B_\parallel\)], which polarizes spins of scatterers without affecting orbital motion of free carriers. Here, we show that in 2D conductors with substantially different values of the g-factors of electrons (\(g_e\)) and magnetic defects (\(g_s\)), the decoherence time [\(\tau_s(B_\parallel)\)] (reflected by the curvature of magneto-conductance) displays an anomaly: it first gets shorter, decaying on the scale [\(B_\parallel \sim \hbar/(|g_s| - |g_e|\mu_B\tau_s)\)], before becoming longer at higher values of [\(B_\parallel\)].

The electron interference results in a quantum correction to the Drude conductivity and a positive magneto-conductivity (MC) of disordered metals. In particular, the constructive interference of electron waves propagating in time-reversed fashion along closed diffusive loops in two dimensional (2D) conductors brings about a logarithmically divergent weak localization (WL) correction. In the absence of external magnetic flux piercing the electron trajectories, this divergence of WL correction is cut off by the electron decoherence time [\(\tau_s\)]. Application of the flux breaks the time reversal symmetry, thus further diminishing the WL correction and leading to the low-temperature MC, \(\sigma(B_\parallel)\), where \(B_\parallel\) is the magnetic field component perpendicular to the plane of a 2D sample. The MC curvature, \(\kappa \equiv \frac{\partial^2 \sigma}{\partial B_\parallel^2}\Big|_{B_\parallel=0} \propto \tau_s^{-2}\), gives a measure for the electron coherence time in 2D conductors: doped semiconductor quantum wells, charge accumulation layers near semiconductor interfaces, thin metallic films, or atomically-thin 2D crystals such as graphene and transition-metals dichalcogenides monolayers.

The two leading decoherence processes at low temperatures stem from the inelastic scattering of electrons off each other and off magnetic impurities. The electron-electron scattering results\(^2\) in the linear temperature dependence of the decoherence rate, \(\tau_s^{-1} = \frac{kT}{\hbar} e^2/h \ln \frac{\sigma}{2e^2/h}\), with \(\tau_s^{-1} \rightarrow 0\) as \(T \rightarrow 0\). In contrast, the contact exchange interaction with paramagnetic defects results in an apparent low-temperature saturation\(^5\) of the electron decoherence rate at \(\tau_s^{-1}(T \rightarrow 0) \sim \tau_s^{-1}\). The rate \(\tau_s^{-1}\) characterizes the electron spin relaxation due to the spin flips in the course of electron scattering off randomly oriented magnetic moments of impurities.

It is a common knowledge\(^13\) that electron spin relaxation may be suppressed and \(\tau_s\) extended by the polarization of magnetic impurities. In 2D conductors, this can be achieved by using an in-plane magnetic field, \(B_\parallel\), which polarizes the spins of the impurities but does not create any flux through the electron orbits. Measurements of \(\kappa\) in various low-dimensional materials\(^13\) have been reported.[13,14] The inset shows the respective contributions to the full MC curve in absence of in-plane magnetic field, see Eq. (12) and text for details. Here, \(T = 0.5\, \text{K}, D = 100\, \text{cm}^2/\text{s}, \) and \(\tau_s = 0.1\, \text{ns}\).
shown a gradual increase of $\tau_{\varphi}(B_\parallel)$ associated with the spin polarization at $g_i \mu_B B_\parallel S \gtrsim T$ (here $g_i$ and $S$ are respectively the $g$-factor and spin of a magnetic impurity; $\mu_B = |e|\hbar/2mc$ is the Bohr magneton).

Here we show that in some 2D materials the dependence of decoherence time on the in-plane magnetic field, $\tau_{\varphi}(B_\parallel)$, may be non-monotonic: the naively expected polarization-induced increase of $\tau_{\varphi}$ with $B_\parallel$ is preceded by its decrease at weak fields, see Fig. 1. This acceleration of decoherence comes from the precession dynamics of localized magnetic moments and requires the $g$-factors of the impurities ($g_i$) and electrons ($g_e$) to differ from each other. To mention, if $g_i = g_e$, then the local moments are static in the frame rotating together with the precessing electron spins; and in this case the $\tau_{\varphi}(B_\parallel)$ dependence remains monotonic, being caused solely by the impurity spin polarization. For $g_i \neq g_e$, electrons witness the landscape of magnetic moments that varies in time with the frequency

$$\Omega_B = \frac{(g_e - g_i) \mu_B B_\parallel}{\hbar}.$$  

(1)

This temporal variation shortens $\tau_{\varphi}$, if $\Omega_B \tau_S \gtrsim 1$. The latter condition is satisfied already at non-polarizing fields, assuming that $|1 - g_i/g_e|kT\tau_S/\hbar S \gg 1$.

Polarization of the magnetic impurities renders them ineffective in the electron phase relaxation, thus leading to a strong increase of the magnetoconductance signal.\textsuperscript{19,20} Contrary to that, the effect of spin dynamics is quite subtle. We find the limitation on the magnitude of the corresponding change in the magnetoconductance, evaluate analytically the dependence of the $\tau_{\varphi}$ on $B_\parallel$, and relate it to the basic parameters of the itinerant electrons and magnetic impurities.

To analyze the influence of spin-flip scattering in a conductor on the WL effect, we consider an electron wave propagating along a closed-loop trajectory, scattering from disorder, $V(r) = \sum_r U(r - r_1) + \sum_j J S_j$, the Zeeman terms for electrons and impurities are $-g_i \mu_B B_\parallel \sigma_z$ and $-g_i \mu_B B_\parallel |S_j|_z$, respectively (z-axis is chosen along the in-plane magnetic field), $\sigma$ is the electron spin operator acting on the $\pm 1/2$ spin states quantized along z-axis.

To quantify the $\tau_{\varphi}(B_\parallel)$ dependence, we express the WL correction $\delta \sigma$ to conductivity\textsuperscript{19} in terms of two-electron propagators, ‘Cooperons’ $C_{\sigma\sigma';\eta\eta'}(\epsilon, q, \omega', \omega)$, see Fig. 2(1):

$$\delta \sigma = \frac{e^2}{2\pi \hbar} [C_{0,0} - C_{1,0} - C_{1,1} - C_{1,-1}];$$

$$C_{S,M} = \zeta_{S,M;\sigma\sigma'} C_{S,M;\sigma\sigma'};$$

$$\hat{C} = -\gamma v_F^2 r^3 \int d\epsilon d^2q n_F'(\epsilon') \hat{C}(\epsilon, q).$$  

(2)

Here Clebsch-Gordan coefficients $\zeta_{S,M;\sigma\sigma'} = \langle S, M | \frac{1}{2}, \frac{1}{2}; \sigma' | S, M \rangle$ select from the Cooperon matrix $\hat{C} = \int d\omega C_{\sigma\sigma';\eta\eta'}(\epsilon, q, \omega, \omega')$ the singlet ($S = 0$) and triplet ($S = 1, M = -1, 0, 1$) components defined in terms of the total spin carried by the two-electron propagator and its projection onto the external magnetic field $B_\parallel$. Also,

$$\epsilon_r = \epsilon - \sigma g_i \mu_B B_\parallel,$$

(3)

and $n_F'(\epsilon) = \partial n_F(\epsilon)/\partial \epsilon$ is a derivative of Fermi distribution function\textsuperscript{21}.

The diagrammatic form of the Bethe-Salpeter equation for the Cooperon matrix $\mathcal{C}$ is shown in Fig. 2(2). Its important element is the disorder correlation function represented by the dashed lines in Fig. 3 which is assumed to be short-range and includes the following elements:

(a) Correlator of spin-less disorder, $n_{UL} U^2 \delta(\omega) \delta(\mathbf{r} - \mathbf{r}')$, where $n_{UL}$ is the density of the point-like potential scatterers;

(b) Correlator of the z-spin components of local magnetic moments that characterizes spin-dependent scattering of electron without spin flip, $n_{J} J^2 T_{K S_z}(t) S_z(t') \delta(\mathbf{r} - \mathbf{r}')$. Here, $n_{J}$ is the density of the magnetic defects of spin $S$. The spin-correlator $\langle T_{K S_z}(t) S_z(t') \rangle$ is independent on the positions $t$ and $t'$ on the Keldysh contour, hence, it has only Keldysh component with the Fourier transform $n_{J} J^2 T_{K S_z}(t) S_z(t') \delta(\mathbf{r} - \mathbf{r}')$.

(c,d) Spin correlators $n_{J} J^2 T_{K S_z}(t) S_z(t') \delta(\mathbf{r} - \mathbf{r}')$, where $D(t,t') = \langle T_{K S_z}(t) S_z(t') \rangle$ is mapped from the

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig2}
\caption{Disorder perturbation theory diagrams for the WL correction to conductivity. (1) WL correction related to the Cooperon $C_{\sigma\sigma';\eta\eta'}(\epsilon, q, \omega', \omega)$. Bold dots stand for the current operators, bold lines are the disorder-averaged Keldysh functions. (2) Bethe-Salpeter equation for the Cooperon. (3) Combination of components of Keldysh functions involved in the kernel of the Cooperon equation. The first diagram in the right part of the equation contains all types of scattering described in Fig. 3(a-d), while the second and third contain the spin-exchanges processes (c,d) only.}
\end{figure}
Keldysh time-contour onto the matrix Keldysh space with components
\[
D^{R/A}(\omega) = 2i\langle S_+ \rangle (\omega - g_i \mu_B B \pm i\delta)^{-1},
\]
\[
D^K(\omega) = 4\pi |S(S + 1) - \langle S_z^2 \rangle| \delta(\omega - g_i \mu_B B).
\]  

Here, \( \langle S_+ \rangle = [Z(a)]^{-1} \partial_\sigma Z(a), a = g_i \mu_B B || /kT \) and \( Z(a) = \sum_{S_{\sigma,\sigma'}} e^{aS_z} \) is the partition function for a paramagnetic scatterer.

Thick solid lines in Fig. 2 stand for disorder-averaged electron Green’s functions \( G_\sigma \), obtained from the solution of the Dyson equation shown in Fig. 3.

\[
G^{R/A}_\sigma = (\varepsilon_\sigma - v_F \xi_\sigma + g_i \mu_B B |\sigma| \pm \frac{i}{2} (\tau^{-1} + \tau^{-1}))^{-1};
\]
\[
G^K_\sigma = (1 - 2n_F(\varepsilon_\sigma))(G^R_\sigma - G^A_\sigma);
\]  

where \( \tau = 1/2\pi\gamma n_{0} U^2 \) is mean free time, \( \tau_s = 1/(2\pi\gamma n_{j} J^2 S(S + 1)) \) is spin relaxation time, \( \gamma \) is the electron density of states,

\[
\tau_\sigma^{-1} = \left[ 1 - 2\sigma M_1(1 - 2n_F(\varepsilon_\sigma)) \right] \tau_s^{-1};
\]
\[
M_1 = \langle S_n^0 \rangle / S(S + 1); \quad \xi_\sigma \approx v_F |p| - p_F,
\]  

and \( \sigma = \pm \frac{1}{2} \) is electron’s spin projection on the direction of in-plane magnetic field.

The spin structure of the Cooperons \( C_{1,\pm 1} \) allows only for the (a) and (b) contributions to the dashed line in the bottom row in Fig. 2, forbidding the spin exchange (c,d), and thus securing \( \mathcal{C}_{\sigma \sigma',\sigma \sigma'}(\varepsilon, q; \omega', \omega) \propto \delta(\omega') \). The kernel of the Bethe-Salpeter equation for Cooperons \( C_{1/0,0} \) includes spin-exchange contributions (c,d). Summing up all three possible combinations of Keldysh functions components \( G^A D^K G^R, G^A D^A G^K, \) and \( G^K D^K G^K \) in Fig. 3, where the frequency argument of the spin correlator is \( D_{R/K/A} \), we get

\[
\frac{n J^2}{2} \left\{ D^K + [1 - 2n_F(\varepsilon_\sigma + \omega)](D^R - D^K) \right\}
= 2\pi n J^2 \left\{ (S(S + 1) - \langle S_z^2 \rangle) - [1 - 2n_F(\varepsilon_\sigma)](\Omega_2) \right\} \delta(\omega).
\]

The frequency dependence of this kernel enforces \( \mathcal{C}_{\sigma \sigma',-\sigma,-\sigma}(\varepsilon, q; \omega', \omega) \propto \delta(\omega') \), i.e. the energy transferred through the impurity spin correlator can be only \( \pm g_i \mu_B B \), where sign depends on whether the spin transferred to the defect is +1, or −1. After taking this into account, the equation for the Cooperon takes the form

\[
\left( D \left( -i\hbar \partial_\tau + \frac{2e}{c} A_\perp \right)^2 + \hat{R} \right) \hat{\mathcal{C}}(\varepsilon, \mathbf{r}) = \frac{1}{2\pi\gamma^2 \tau^2} \delta(\mathbf{r});
\]

\[
R_{\sigma \sigma',\sigma'\sigma}(\varepsilon, q; \omega', \omega) = \delta(\varepsilon_\sigma - \varepsilon_\sigma') \delta(\sigma - \sigma') \left[ (\tau_\sigma^{-1} + \tau_{\sigma'}^{-1})/2 + \tau_T^{-1} \right] 
+ \delta(\sigma - \sigma') \delta(\sigma + \sigma') \left[ -4\sigma\sigma'\tau_s^{-1} M_2 + |\sigma - \sigma'| \tau_s^{-1} + i(\sigma - \sigma') \Omega_B \right],
\]  

where \( D = v_F^2 \tau/2 \) is the diffusion coefficient, \( \hat{\mathcal{C}}(\varepsilon, \mathbf{r}) \) is the Fourier transform of \( \hat{\mathcal{C}}(\varepsilon, q) \) and \( A_\perp = (0, B_\perp x) \) is the vector potential of the perpendicular magnetic field (note that \( B_\perp \ll B || \)).

Diagonalization of a matrix \( \hat{R} \) produces Cooperons \( C_{1,\pm 1} \) decoupled from all other Cooperon components and having decay rates

\[
\tau_1^{-1} \pm 1 = [1 - M_2 \mp M_1(1 - 2n_F(\varepsilon_\sigma))] \tau_s^{-1} + \tau_T^{-1}.
\]  

Here \( \varepsilon_\pm \), see Eq. 3, accounts for the energy transfer to an impurity in the process of spin-flip scattering. The Cooperon components \( C_{0,0} \) and \( C_{1,0} \) are coupled with each other by spin-flip processes. The coupling generates combined modes decaying with the rates

\[
\tau_0^{-1} = \left[ 1 + M_2 - M_1(\varepsilon_+ - n_F(\varepsilon_-)) \right] \tau_s^{-1} \pm \sqrt{\tau_{1,1}^{-1} \tau_{1,-1}^{-1} - \Omega_B^2 + \tau_T^{-1}}.
\]
Note that at $B_{\parallel} = 0$ the average values $\langle S_\parallel \rangle = 0$ and $\langle S_\perp^2 \rangle = \frac{1}{3} S (S + 1)$, so that $\tau_{1,\pm,1}^{-1} = \tau_{0,1}^{-1} = \frac{2}{3} \tau_s^{-1} + \tau_T^{-1}$ and $\tau_{0,\pm}^{-1} = 2 \tau_s^{-1} + \tau_T^{-1}$, in agreement with earlier theories.

Relaxation rates $\tau_{1,\pm}^{-1}$ and $\tau_{0,\pm}^{-1}$ vary with $B_{\parallel}$ over two parametrically different field scales. For $\tau_{1,\pm,1}$, the scale is determined by the polarization of impurity spins. The polarization takes place at $g_i \mu_B B_{\parallel} S \gg kT$, and Eq. (3) then yields $\tau_{1,\pm,1}^{-1} \sim \tau_T^{-1} \ll \tau_s^{-1}$. On the contrary, the field dependence of $\tau_{0,\pm}^{-1}$ is defined by the electron and impurity spin dynamics. Under the condition $|1 - g_e/g_i| kT \tau_s/hS \gg 1$, the corresponding field scale is much smaller. Neglecting the spin polarization, we may simplify Eq. (9) to:

$$\tau_{0,\pm}^{-1} = \frac{4}{3} \tau_s^{-1} \pm \sqrt{\left(\frac{2}{3} \tau_s^{-1} - \Omega_B^2 + \tau_T^{-1}\right)^2}.$$  \hspace{1cm} (10)

As expected, the effect of the magnetic field depends on $\Omega_B$, the difference between the precession frequencies of the impurity and electron spins, see Eq. (1). The effect is absent if the corresponding g-factors are identical.

When substituted in Eqs. (2), the four Cooperon modes obtained using Eq. (4) yield the WL correction to the conductivity at $B_{\parallel} = 0$ (the first term in square brackets comes from $C_{1,\pm,1}$ and the second from $C_{1,0,0}$),

$$\delta \sigma = \frac{e^2}{2\pi h} \int d\varepsilon \sum_{\alpha=\pm} n_F'(\varepsilon_\alpha) \left[ \ln \frac{\tau_{0,\pm}}{\tau_{\min}} + A_0 \ln \frac{\tau_{0,\pm}}{\tau_0} \right] + A_\pm = (\tau_{1,\pm,1}^{-1} - \tau_T^{-1})/(\tau_{0,\pm}^{-1} - \tau_{0,\pm}^{-1}).$$  \hspace{1cm} (11)

Here, the ultraviolet cutoff under the logarithm is, typically, $\tau_{\min} \sim \tau$, but for the description of WL effect in graphene, $\tau_{\min}$ one should use for $\tau_{\min}$ in Eq. (11) the intervalley scattering time $\tau_{\text{iv}}$, instead of mean free path time $\tau$. The MC, studied as a function of $B_{\parallel}$ for fixed $B_{\perp}$, takes the form

$$\sigma(B_{\perp}, B_{\parallel}) - \sigma(0, B_{\parallel}) = -\frac{e^2}{2\pi h} \int d\varepsilon \sum_{\alpha=\pm} n_F'(\varepsilon_\alpha)
\times \left\{ \frac{F(B_{\parallel}/B_{1,\alpha}) + F(B_{\parallel}/B_{0,\pm}) - F(B_{\parallel}/B_{0,\pm})}{A_0} \right\} + F(z) = \ln z + \psi\left(\frac{2}{z}\right) + \frac{\bar{B}_{\beta,\alpha} = h/4e}{D \tau_{\beta,\alpha}},$$  \hspace{1cm} (12)

where $\psi$ is digamma function.

One may see that the part stemming from $C_{1,0,0}$ (square brackets) saturates at high $B_{\parallel}$; it contributes (in units of $e^2/2\pi h$) at most $(3/4) \ln 3 \approx 0.82$ to the MC. This should be contrasted with the logarithmic growth with $B_{\parallel}$ of the term coming from $C_{1,\pm,1}$ (first term in braces). That peculiarity of the field dependence sets the dynamic range of MC useful for extracting the MC curvature, $\kappa(B_{\parallel}) \propto \tau_{\perp}^2$, using the expansion $F(z) \approx z^2/24 + O(z^3)$, as illustrated in the inset of Fig. 1. In the ‘high-temperature’ limit,

$$kT \gtrsim hS/\tau_s [1 - g_e/g_i],$$  \hspace{1cm} (13)

the expression for $\kappa$ can be simplified further for all of $B_{\parallel}$, if inelastic e-e collisions are neglected ($\tau_T^{-1} \to 0$),

$$\kappa \approx \frac{e^2}{\pi h} \left( \frac{D \tau_{\beta}}{h/e} \right)^2 \left\{ 2 W_B + \frac{4/3}{(1+4\Omega_B^2/T_s^2)} \right\},$$

$$W_B = ch(2a) - ch(a), \quad a = g_i \mu_B B_{\parallel}/kT,$$  \hspace{1cm} (14)

$M_1 = (S + 1/2) \coth((S + 1/2)a) - 1/2 \coth(a)$, $S(S + 1)$ where $(S + 1)M_1$ is Brillouin function [see Eq. (3)]. The first term in brackets in Eq. (14) comes from Cooperons $C_{1,\pm,1}$ of Eq. (2) and has two asymptotes: $3 + 29 + 8S(1 + S)$ at $B_{\parallel} \ll kT/g_i \mu_B S$ and $1/(S + 1)^2 \exp(2g_i \mu_B B_{\parallel}/kT)$ at $B_{\parallel} \gg kT/g_i \mu_B$. The latter exponential asymptote is cut off by inelastic e-e scattering resulting in $\kappa = 2e^2/(3\pi h) \tau_{\perp}^2$. The second term in brackets originates from $C_{1,0,0}$ and its contribution to $\kappa(B_{\parallel})$ decays with increasing $B_{\parallel}$. Together, the two contributions provide the non-monotonic dependence of magnetococonductance curvature (which is conventionally considered as the measure of coherence time) over the in-plane field scale $B_{\parallel} \sim h/(g_i - g_e) \mu_B T$. This non-monotonic dependence includes a local maximum at $B_{\parallel} = 0$ and a minimum at $B_{\parallel} \sim kT/g_i \mu B S^2 (1 - g_i/g_e)^{-2/3}$, which is followed by the increase of $\kappa(B_{\parallel})$ due to the polarization of defect spins, cf. Eqs. (13) and (14). Eventually MC curvature saturates at the scale set by the inelastic electron-electron scattering decoherence time $\tau_T$.

The above-described anomalous behavior of decoherence rate occurs only when the electron orbit/and magnetic defect have g-factor values different from the free-electron $g = -2$. The values of $g_i \neq -2$ may be caused by the crystalline anisotropy effect on a heavy ion embedded in a 2D metal or semiconductor ($g_a$, graphene). For example, crystalline anisotropy splits states of a spin $1/2$ magnetic ion into two Kramers doublets with spin projections $\pm 1/2$ and $\pm 3/2$ onto the direction perpendicular to the plane of the 2D electron system. Then, the Zeeman splitting by the in-plane magnetic field realizes the cases of $g_i = -4$ (yellow line in Fig. 1) and $g_i = 0$ (orange line in Fig. 1) for the two doublets, respectively. The case of $g_e = -2, g_i \neq -2$ was realized in graphene exfoliated on a SiO$_2$/Si substrate where the non-monotonic magnetococonductance is well-described by the presented here theory. Alternatively, the situation $g_i = 0$ (green line in Fig. 1) can appear in p-doped transition metal dichalcogenides MoS$_2$, MoSe$_2$, WS$_2$ or WSe$_2$, where, due to a large spin-orbit splitting, Kramers doublets of the hole states correspond to opposite spins in the opposite valleys, and the external in-plane magnetic field does not lift the Kramers degeneracy.

In conclusion, the difference in the precession frequencies of the electron and impurity spins results in a non-monotonic dependence of the electron decoherence time to
on the magnetic field causing the precession. We find the magnitude and the functional form of that dependence, and relate it to the parameters of the itinerant electrons and magnetic impurities. Despite being small, the effect is important, as a manifestation of the very basic physics of magnetic moments in solids.

**ACKNOWLEDGMENTS**

We thank I. Aleiner, B. Altshuler and M. Vavilov for useful discussions. This work has been supported by the Deutsche Forschungsgemeinschaft and Research Training Group GRK 1621, ERC Synergy Grant Hetero2D, Royal Society, and US NSF DMR-1206612.