Weak localization effects in granular metals

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The weak localization correction to the conductivity of a granular metal is calculated using the diagrammatic technique in the reciprocal grain lattice representation. The properties of this correction are very similar to the corresponding one in disordered metal, with the replacement of the electron mean free path \(\ell\) by the grain diameter \(d\) and the dimensionless conductance \(g\) by the tunnelling dimensionless conductance \(g_T\). In particular, we demonstrate that at zero temperature no conducting phase can exist for dimensions \(D \leq 2\). We also analyze the WL correction to magnetoconductivity in the weak field limit.

Recently, the properties of granular materials have attracted special attention. The quantization of the electron spectrum in small grains requires to revise the basic idea of the quasiparticle spectrum continuity, assumed in the description of most properties of metallic and superconducting systems. The appearance of a new energy scale, the mean level spacing \(\delta\), results in unusual superconducting properties of such systems, possibility to observe there specific quantum phase transitions, etc.

In particular, it turns out that the interplay between the intra-grain diffusion and inter-grain tunnelling of electrons makes the metal-insulator transition in such “quantum metal” very peculiar. In this Letter, we intend to discuss the specifics of the weak localization corrections in granular systems.

The elastic electron relaxation rate in granular metal consists of three contributions

\[
\frac{1}{\tau_{el}} = \frac{1}{\tau_{imp}} + E_T + \Gamma,
\]

where \(\tau_{imp}\) is the mean scattering time of electrons with impurities, \(E_T = v_F/d\) is the Thouless energy, \(d\) is the characteristic grain size, \(v_F\) is the intra-grain Fermi velocity. The last term is the electron inter-grain tunnelling rate: \(\Gamma = g_T \delta\), where \(g_T = 2\pi (t/\delta)^2\) is the tunnelling dimensionless conductance and \(t\) is the tunnelling energy.

The character of electron motion in a metal is conveniently classified as a function of its mean free path \(\ell\). The diffusion length \(\ell_{n}\) is \(\ell_{n} = \sqrt{D_n/T}\) (where \(D_n = v_F^2 \tau_{imp}/D\) is the diffusion coefficient of metal and \(D\) is the space dimensionality) separates the regions of ballistic (\(\ell \gg \ell_T\) and diffusive (\(\ell \ll \ell_T\)) electron motion. When \(\ell \to h/p_F\) the metal-insulator transition in 3D case takes place. In the case of a granular metal, one can expect that the standard WL theory, describing the precursor effects of this transition, has to be modified in two ways. First, the diffusion coefficient \(D\) here, at least in some interval of parameters, has to be determined by the tunnelling time \(\Gamma^{-1}\) instead of elastic scattering time \(\tau_{imp}\) and therefore we will have a different tunnelling thermal length \(\ell_T = \sqrt{D/T}\). Second, the characteristic grain size \(d\) must appear in the theory side by side with the diffusion length \(\ell_T\). Two different situations are possible. When \(d \ll \ell_T\), the diffusive part of the \(\ell\)-axis is divided to the regions of the normal intra-grain diffusion (\(h/p_F \ll \ell \ll d\)) with the metallic diffusion coefficient \(D_n\) and of the tunnel inter-grain electron diffusion (\(d \ll \ell \ll \ell_T\)) characterized by the diffusion coefficient \(D = \left(\langle x(t)^2\rangle/t\right)_{t \to \infty} \approx \Gamma d^2\) (see Fig. 1b). In the opposite limit, when \(d \gg \ell_T\), only the intra-grain diffusion is possible, but, in its turn, the domain of ballistic regime contains two regions: the intra-grain one with ballistic time \(\tau_{imp}\) (\(\ell_T \ll \ell \ll d\)) and the region of inter-grain electron motion with the ballistic time \(\Gamma^{-1}\) (see Fig. 1b).

We will discuss here the most interesting case of low temperatures, \(d \ll \ell_T\). We will also assume that the electron motion inside a single grain is ballistic (\(\ell_{bulk} \gtrsim d\), \(\ell \gg d\)): this means that before tunnelling to the neighbor grain the electron is reflected many times on the grain boundaries (\(\Gamma \ll E_T\)). As a consequence, \(\tau_{el} \approx E_T^{-1}\)

![FIG. 1: A schematic representation of the two possible scenarios for the electron motion in granular metals as a function of the ratio between \(d\), the grain diameter, \(\ell_T\), the thermal length, and \(\ell\), the mean free path.](image)
and the dimensionless conductance of a single grain \( g = E_T / \delta \gg g_T \). The conductance of the entire system is given by \( g^{-1} + g_T^{-1} = g_T^{-1} \), what is equivalent to say that the drop of applied electric potential occurs only inside the tunnel barrier.

We assume that the grains are almost identical, with average diameter \( d \), and form a regular lattice with lattice constant equal to the same \( d \). The coordinate of the grain center will be labelled by the lattice variable \( \mathbf{R} \). The Hamiltonian of the system can be written as \[ 1 \]

\[
\hat{\mathcal{H}} = \sum_{i,\mathbf{p}} \varepsilon_i \hat{c}^\dagger_{i,\mathbf{p}} \hat{c}_{i,\mathbf{p}} + \frac{1}{2} \sum_{\langle i,j \rangle, \mathbf{p}, \mathbf{p}'} \left[ \sum_{\mu} \left[ \hat{c}^\dagger_{i,\mathbf{p}} c^\dagger_{j,\mathbf{p}'} + h.c. \right] \right] \gamma_{i,j} \]

where \( \hat{c}_{i,\mathbf{p}} \) (\( \hat{c}_{i,\mathbf{p}}^\dagger \)) is the annihilation (creation) operator of an electron in grain \( i \) with intra-grain momentum \( \mathbf{p} \). The tunneling energy \( \gamma_{i,j} \) will be taken equal for all bonds between nearest neighbor grains and independent on the intra-grain momentum, \( \gamma_{i,j} = t \). Performing the Fourier transform with respect to such \( \mathbf{R} \), one can write the Hamiltonian in the representation of both intra- and inter-grain momenta (double momentum representation):

\[
\hat{\mathcal{H}} = \sum_{\mathbf{K}, \mathbf{p}} \left[ \varepsilon_{\mathbf{p}} + t Z \gamma_{\mathbf{K}, \mathbf{p}} c^\dagger_{\mathbf{K}, \mathbf{p}} c_{\mathbf{K}, \mathbf{p}} + \frac{t Z}{2} \sum_{\mathbf{K}, \mathbf{p}, \mathbf{p}'} \gamma_{\mathbf{K}, \mathbf{p}, \mathbf{p}'} \left[ c^\dagger_{\mathbf{K}, \mathbf{p}} c^\dagger_{\mathbf{K}, \mathbf{p}'} + h.c. \right] \right]. \tag{2}
\]

Here \( \mathbf{K} \) is the quasi-momentum belonging to the reciprocal to \( \mathbf{R} \) lattice and it varies in the first Brillouin zone. The lattice structure factor \( \gamma_{\mathbf{K}, \mathbf{p}} = Z^{-1} \sum_{\mu=1}^{Z} e^{i \mathbf{K} \cdot \mathbf{d}_\mu} \) are the vectors connecting the center of selected grain with the nearest neighbor sites, \( Z \) is the coordination number. For a simple cubic lattice, the vectors \( \mathbf{d}_\mu \) have one component equal to \( \pm d \) and all the others equal to zero. For sake of simplicity, we will restrict to this case; the extension to generic lattices is straightforward. In simple cubic lattices, \( \gamma_{\mathbf{K}, \mathbf{p}} = \frac{1}{2} \sum_{\mu=1}^{D} \cos(K_\mu \xi) \). From Eq. \( 2 \) we can define the single electron Green’s function in the double momentum representation as:

\[
G_{\mathbf{K}}(\mathbf{p}, \varepsilon_n) = \frac{1}{i \varepsilon_n - \varepsilon_{\mathbf{p}} - Z t (1 - \gamma_{\mathbf{K}, \mathbf{p}})}, \tag{3}
\]

with \( \varepsilon_n = \varepsilon_n + (2 \tau_d)^{-1} \text{sgn} \varepsilon_n \) and \( \varepsilon_n = \pi T (2n + 1) \) as fermionic Matsubara’s frequency.

Recalling that the electric field is negligible inside the grains and differs from zero only inside the barriers, in presence of the vector potential \( \mathbf{A} \) one can write the \( \alpha \)-th component of the electrical current operator in the imaginary time \( \tau \) as:

\[
\hat{J}_\alpha(\tau) = \frac{e d}{2} \sum_{\mathbf{K}, \mathbf{p}, \mathbf{p}'} \left[ e^{i \mathbf{K} \cdot \mathbf{d}} c^\dagger_{\mathbf{K}, \mathbf{p}}(\tau) \hat{c}^\dagger_{\mathbf{K}, \mathbf{p}'}(\tau) - h.c. \right] \varepsilon_{\mathbf{p}} - \frac{e^2 d}{2} \mathbf{A}(\tau) \cdot \mathbf{d} \hat{H}_T(\tau). \tag{4}
\]

The linear response function, expressed as the second derivative of the partition function, is given by:

\[
\Pi_{\alpha,\alpha'}(\tau) = -\frac{1}{Z[0]} \left. \delta^2 Z[A] \right|_{A=0}^{\delta A_{\alpha}(\tau) \delta A_{\alpha'}(0)} = \Pi_{\alpha,\alpha'}(\tau) - 2 e^2 d^2 \delta_{\alpha,\alpha'} \delta(\tau) \hat{H}_T(0), \tag{5}
\]

where the current-current correlation function is expressed via the current operator as:

\[
\Pi_{\alpha,\alpha'}(\tau) = -\left. \langle \hat{T}_\tau \hat{J}_\alpha(\tau) \hat{J}_{\alpha'}(0) \rangle \right|_0.
\]

The thermal average \( \langle \ldots \rangle_0 \) shall be performed with the diagonal Hamiltonian, the first line of Eq. \( 2 \). We are interested in the diagonal components of conductivity tensor:

\[
\Pi_{\alpha,\alpha}(\omega) = 2 e^2 d^2 |t|^2 \sum_{\mathbf{K}} \sin^2(K_\alpha d) \times T \sum_{\varepsilon_n \mathbf{p}, \mathbf{p}' \mathbf{p}'} G_{\mathbf{K}}(\mathbf{p}, \varepsilon_{n+\nu}) G_{\mathbf{K}}(\mathbf{p}', \varepsilon_n), \tag{6}
\]

where \( \varepsilon_{n+\nu} = \varepsilon_n + \omega_{\nu} \) and \( \omega_{\nu} = 2 \pi T \nu \) is the bosonic Matsubara’s frequency. We can formulate the following rules of diagrammatic technique in the double momentum representation: 1. at each external vertex attach a factor \( \hat{v}_\alpha = e t d \sin(K_\alpha d) \); 2. at each straight line attach a single electron Green’s function \( G_{\mathbf{K}}(\mathbf{p}, \varepsilon_n) \); 3. sum over all internal momenta and Matsubara’s frequencies; 4. impose energy and lattice momentum conservation at each vertex. Now the impurity averaging of Eq. \( 6 \) can be performed. The Cooperon vertex \( \Pi_0 \) corresponding to the granular metal can be obtained from the Dyson’s equation reported in Fig. \( 2 \). Other possibility is to renormalize the standard intra-grain Cooperon by means of introduction in the corresponding diagrams of a self-energy correction appearing due to tunnelling, as it is done in Ref. \( 1 \). Both approaches in the assumptions made above turn out to be completely equivalent and lead to the expression:

\[
\mathcal{C}_Q(\omega) = \frac{1}{2 \pi T e d \mu F} |\omega| + \frac{1}{2 \Gamma (1 - \gamma Q)}.
\]
In the latter expression appears the exact value for the tunnelling rate: $\Gamma = Z g_T \delta$. In the expression $D = \left[2\Gamma (1 - \gamma_Q) / |Q|^2 \right]_{Q \to 0} = \Gamma^2$ one can also recognize the effective "tunnelling diffusion constant".

Now one can directly calculate the diagram reported in Fig. 3. In our assumptions, see Eq. (4), the Green’s function can be considered independent on $K$ when integrating over $p$, because its behavior is completely determined by the pole due to the impurity (or grain boundaries) scattering, related to $\tau_d^{-1}$, just as in the case of the diffusive limit for a bulk system. Performing the frequency summation, the $p$ and $p'$ integration and the sum over the lattice momentum $K$ one finds

$$K^{WL}_{\alpha \alpha} (\omega) = -\frac{e^2 d^2}{4\pi^2} g_T \sum_Q \frac{\cos (Q_\alpha d)}{|\omega_i| + 2\Gamma (1 - \gamma_Q)}$$

At this point, we can find the WL correction to the conductivity as

$$\frac{\delta \sigma^{WL}_{(D)}}{\sigma^{(D)}} = -\frac{1}{\pi g_T} \sum_Q \frac{\cos (Q_\alpha d)}{1 - \gamma_Q}.$$ (7)

In the case of a bulk granular system ($D = 3$) the sum converges and the correction is finite. The metal-insulator phase transition can be observed at a critical value $Z g_T^2 \approx O(1)$.

In the case of granular film or wire ($D \leq 2$) the WL correction Eq. (7) diverges at small $Q \to 0$. This fact indicates that at zero temperature these systems cannot exhibit metallic properties for any value of the dimensionless conductance $g_T$. At finite temperatures, a natural cut-off of Eq. (7) is given by the phase-breaking rate $\gamma_T = (\Gamma \tau_T)^{-1} = (d/L_T)^2$. It is related to the phase-coherence length $L_T = \sqrt{D \tau_T}$, which tends to infinity when $T \to 0$. In result

$$\frac{\delta \sigma^{WL}_{(1)}}{\sigma^{(1)}} \approx -\frac{2}{Z \pi^3 g_T} \sqrt{\gamma_T} \frac{1}{\gamma_T}, \quad \frac{\delta \sigma^{WL}_{(2)}}{\sigma^{(2)}} \approx -\frac{1}{Z \pi^2 g_T} \ln \frac{\pi^2}{\gamma_T}.$$ (8)

Eqs. (8) permit to define the localization length $\xi^{loc}_{(D)}$ at which the correction becomes of the order of $\sigma^{loc}_{(D)}$:

$$\frac{\xi^{loc}_{(1)}}{d} \approx \frac{Z \pi^3}{2 g_T} \frac{\xi^{loc}_{(2)}}{d} \approx \frac{Z \pi^2}{2 g_T}.$$

It is worth to stress that Eq. (7) does not contain $\tau_d$ but only lattice parameters as $g_T$ and the coordination number $Z$: the intra-grain dynamics, as expected, simply drops out of the calculation. The conductivity and its corrections are related to the diffusion on the grain lattice, and the mechanism of the momentum randomization between different grains is not crucial: the electron dynamics at low temperature can be thought as that of a random walker on a lattice. This picture is fully consistent with the existing WL theory [3, 4], and it is in agreement with previous experimental findings in granular metals [8].

More intriguing are the properties of the WL correction to the magnetoresistance of granular metal. It is well known that quantum corrections to conductivity in disordered metal are very sensitive to the magnetic field: in fact, its presence disturbs the phase coherence of electrons moving along the self-intersecting trajectories, suppressing the WL correction and leading to the appearance of the anomalous negative magnetoresistance [6]. In the following, we will show how such a correction manifests itself in the case of granular metal.

To calculate the WL contribution to magnetoresistance, it is necessary to rewrite Eq. (7) in the direct space:

$$\frac{\delta \sigma^{WL}_{(D)}}{\sigma^{(D)}} = -\frac{2}{Z g_T} \frac{\tilde{C}_{i,i+\alpha} + \tilde{C}_{i+\alpha,i}}{2}$$

which is independent on $i$, depending only on the inter-grain spacing $d$. $\alpha$ represents the bond along the direction of the current. Here $\tilde{C}_{i,i}^{-1} = (2\pi)^2 \nu_F C_Q^{-1} = -i\omega + 2\Gamma (1 - \gamma_Q)$. Let us notice that this form underlines the fact that transport is due only to the potential drop inside the barrier separating two grains $i$ and $i + \alpha$. In the presence of a magnetic field, the Cooperon wavefunction is given by the solution of the equation

$$(4\Gamma) (1 - \gamma_{Q+2eA}) \psi_i (r) = E \psi_i (r);$$ (9)

Moreover, also the intra-grain Cooperon will be renormalized by the presence of the magnetic field, acquiring a mass term equal to $\tilde{E}_0 (H) = \frac{\phi}{\phi_0} \left(\frac{\phi}{\phi_0}\right)^2 E_T$, where $\phi = H d^2$ is the magnetic flux threaded through a single spherical grain and $\phi_0 = \pi/e$ is the flux quantum. When the field satisfies the inequality $d \ll \ell_H = (\epsilon H)^{-1/2}$, or $\phi \ll \phi_0$, we have

$$\tilde{C}_{ij} (r, r', \omega) = \sum_{Q_i, Q_j} -i\omega + \Omega_c \left(n + \frac{1}{2}\right) + D Q_i^2 + \frac{1}{\tau} + E_0 (H)$$

$$= \sum_{Q_i, Q_j, n} \psi_{i, Q_i, n} (r) \psi^*_{j, Q_j, n} (r') + Q_i^2 + \frac{1}{\tau} + E_0 (H)$$

FIG. 3: WL correction to the conductivity in the double momentum representation. The solid lines are single electron temperature Green’s functions $G_K (p, \varepsilon_n)$; the external velocity vertices are $\dot{v}_i = etd \sin (K_i d)$; the shaded box is the Cooperon $C_Q (\varepsilon_n)$.
where \( \Omega_c = 4D/\ell_T^2 \) is the Cooperon cyclotron energy. \( Q_\parallel \) is the momentum along the magnetic field and \((n, Q_\perp)\) are the quantum numbers of the Landau basis.

The most interesting case is the two-dimensional geometry, with the magnetic field applied across the plane and \((n, Q_\parallel, Q_\perp)\) are the quantum numbers of the Landau basis. The most singular correction reduces to

\[
\frac{\delta \sigma_{\text{WL}}}{\sigma_0} = \frac{1}{\pi \Delta_{\text{tr}}^2} \sum_{n=0}^{\max_{\Delta_{\text{tr}}} \phi \phi_0, \gamma \phi_0} \left[ \cos \left( \frac{\phi \phi_0}{4\phi_0} \right) + \frac{\pi \phi_0}{10 \phi_0} \frac{E_T}{\Gamma} \right] - \frac{\pi \phi_0}{10 \phi_0} \left[ \cos \left( \frac{\phi \phi_0}{4\phi_0} \right) + \frac{\pi \phi_0}{10 \phi_0} \frac{E_T}{\Gamma} \right] \]

where \( \psi(x) \) is the digamma function. The WL correction to the magnetoresistance \( \delta \sigma \) is

\[
\delta \sigma (H) = \delta \sigma_{\text{WL}} (H) - \delta \sigma_{\text{WL}} (0) \]

One more energy scale shows up in Eq. (12) with respect to the bulk case, namely the Thouless energy. In the limit of very weak fields, \( \phi / \phi_0 \ll \sqrt{1/4E_T \tau_\phi} \), this energy scale is not observable in the magnetoresistance: the leading singular correction reduces to

\[
\frac{\delta \sigma}{\sigma_0} \approx \frac{2\pi}{3} \frac{1}{3 \Delta_{\text{tr}}^2} \left( \frac{\phi}{\phi_0} \right)^2 \propto H^2
\]

which corresponds to the anomalous magnetoresistance of the standard theory. The granular behavior deviates from the bulk one at fields such that \( \phi / \phi_0 \sim \Gamma / E_T \) where the intra-grain term starts to dominate in the second digamma function in \( \delta \sigma (H, \gamma \phi) \). For \( \phi / \phi_0 \ll \gamma \phi \), the magnetoresistance correction acquires the logarithmic form

\[
\frac{\delta \sigma}{\sigma_0} = \frac{2}{\pi \Delta_{\text{tr}}^2} \phi_0 \ln \left( \frac{E_T \phi_0}{\Gamma \phi_0} \right).
\]

Larger fields are out of the range of our approximated approach, in which the intra-grain Landau levels, with the spectrum \( \omega_c (n + 1/2) / \omega_c = 4D_\parallel / \ell_T^2 \), start to significantly contribute to the Cooperon wavefunction. Let us notice finally that the ultraviolet cut-off \( \max_{\Delta_{\text{tr}}} \approx E_T / \omega_c = \Gamma / \Omega_c \) remains the same.

In summary, we developed a diagrammatic technique in a double-momentum representation for transport in granular metals. Using this technique, the weak localization corrections to the conductivity arise in a natural way and an explicit calculation shows the same low-temperature behavior as in bulk metals, but with the diffusion constant \( D_\parallel \) replaced by the effective tunnelling diffusion constant \( D \parallel = \Gamma d^2 \) and the mean free path \( \ell \) by the average grain diameter \( d \). Our result agrees with Eq. (11) of Ref. [4] in the \( Q \to 0 \) limit; however, our technique underlines the presence of the grain lattice, represented by the cosine factor in Eq. (11), reminiscent of the lattice structure factor \( \gamma_Q \). We also give an estimate of the magnetoresistance correction for very weak fields.

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