

Negative Magnetoresistance of Granular Metals in a Strong Magnetic Field

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The magnetoresistance of a granular superconductor in a strong magnetic field that destroys the gap in each grain is considered. It is assumed that the tunneling between grains is sufficiently large such that all conventional effects of localization can be neglected. A nontrivial sensitivity to the magnetic field comes from superconducting fluctuations leading to the formation of virtual Cooper pairs and reducing the density of states. At low temperature, the pairs do not contribute to the macroscopic transport but their existence can drastically reduce the conductivity. Increasing the magnetic field destroys the fluctuations, which improves the metallic properties and leads to the negative magnetoresistance. [S0031-9007(99)08995-4]

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In a recent experiment [1], the resistance of a system of Al grains as a function of the magnetic field was studied. The samples were quite homogeneous with a typical diameter of the grains $120 \pm 20 \text{ \AA}$, and the grains formed a three-dimensional array. By destroying the superconductivity by the magnetic field, we observed that in a sufficiently strong magnetic field the system had a finite resistivity. The applied magnetic fields reached 17 T, which was more than sufficient to also destroy the superconducting gap in each grain.

The dependence of the resistivity on the magnetic field observed in Ref. [1] was not simple. At strong magnetic fields, the resistivity *increased* when decreasing the magnetic field. The resistivity decreased only at sufficiently weak magnetic fields and the samples finally displayed superconducting properties. Similar behavior was reported in a number of publications [2,3]. A negative magnetoresistance due to weak localization effects is not unusual in disordered metals [4]. However, the magnetoresistance of the granulated materials is quite noticeable in magnetic fields exceeding 10 T, which is many orders of magnitude higher than the typical values relevant for the weak localization.

The aim of this Letter is to demonstrate that the magnetoresistance of a granulated metal in a strong magnetic field and at low temperature *must be negative due to superconducting fluctuations*. We consider a system of granules coupled to each other. The tunneling amplitude between the grains is assumed to be large, such that the system without electron-electron interactions would be a good metal macroscopically and would not be sensitive to a magnetic field. The superconducting gap in each granule is assumed to be suppressed by the strong magnetic field. All of the interesting behavior considered below originates from the superconducting fluctuations that lead to a suppression of the density of states (DOS) but do not help to carry an electric current.

The theory of superconducting fluctuations near the transition into the superconducting state was developed long

ago [5–7] (for a review, see Ref. [8]). Above the transition temperature T_c , nonequilibrium Cooper pairs are formed and a new channel of charge transfer opens [5]. Another fluctuation contribution comes from a coherent scattering of the electrons forming a Cooper pair on impurities [6]. Both of the fluctuation corrections increase the conductivity and lead to a positive magnetoresistance. Formation of the nonequilibrium Cooper pairs also results in a fluctuational gap in the one-electron spectrum [7], but in conventional superconductors the first two mechanisms are more important and the conductivity increases when approaching the transition. A small decrease of the transverse conductivity is possible in layered materials [9] in a temperature interval that is not very close to the transition. It is relevant to emphasize that the study of the fluctuations has been done near the critical temperature T_c in a zero or weak magnetic field.

In granulated materials, the superconducting gap in each granule can be destroyed at low temperature by a strong magnetic field. At magnetic fields H exceeding the critical field H_c , virtual Cooper pairs can still be formed. However, as will be shown below, the influence of these pairs on the macroscopical transport is drastically different from that near T_c . The existence of the virtual pairs leads to a reduction of the DOS but, in the limit $T \rightarrow 0$, these pairs cannot travel from one granule to another. As a result, the conductivity σ can be at $H > H_c$, considerably lower than conductivity σ_0 of the normal metal without an electron-electron interaction. It approaches the value σ_0 only in the limit $H \gg H_c$, when all the superconducting fluctuations are completely suppressed by the magnetic field.

For explicit calculations we assume that the superconducting pairing inside the grains is destroyed mainly by the orbital mechanism. The Zeeman splitting is not important provided the radius R of the grains is large enough, so that $R \gg \xi(\epsilon_0\tau)^{-1}$, where ξ is the superconducting coherence length, ϵ_0 is the Fermi energy, and τ is the elastic mean free time. This condition is well satisfied in grains with

$R \sim 100 \text{ \AA}$ studied in Ref. [1]. This limit is opposite to the one considered recently in Ref. [10], where the Zeeman splitting was assumed to be the main mechanism of destruction of the Cooper pairs.

We assume that electrons can hop from grain to grain, and the tunneling energy t is in the interval

$$\delta \ll t \ll \Delta_0, \quad (1)$$

where $\delta = (\nu_0 V)^{-1}$ is the mean level spacing in a single granule, $\nu_0 = m p_0 / 2\pi^2$ is the DOS of the metal in the absence of interactions, V is the volume of the granule, and Δ_0 is the Bardeen-Cooper-Schrieffer (BCS) gap at $T = 0$ in the absence of a magnetic field. The left inequality in Eq. (1) guarantees that the system is macroscopically a good metal [11] and localization effects can be neglected. Moreover, charging effects are also not important in this limit. In the limit $R \ll \xi$ considered below, the superconducting fluctuations in a single grain are effectively zero dimensional (0D).

The Hamiltonian \hat{H} of the system can be written as

$$\hat{H} = \hat{H}_0 + \hat{H}_T, \quad (2)$$

where \hat{H}_0 is a conventional Hamiltonian with an electron-phonon interaction in the presence of a strong magnetic field. The term \hat{H}_T in Eq. (2) describes the tunneling from grain to grain and has the form (see, e.g., Ref. [12])

$$\hat{H}_T = \sum_{i,j,p,q} t_{ijpq} a_{ip}^\dagger a_{jq} + \text{H.c.}, \quad (3)$$

where a_{ip}^\dagger (a_{ip}) are creation (annihilation) operators for an electron, the grain i , and state p .

Correspondingly, the ac current density $\mathbf{j}(t)$ is written using linear response formulas as

$$\mathbf{j}(t) = i \int_{-\infty}^t \langle [\hat{\mathbf{j}}(0, t), \hat{\mathbf{j}}(0, t')] \rangle \mathbf{A}(t') dt', \quad (4)$$

where $\hat{\mathbf{j}}$ is the tunneling current operator and the angle brackets stand for averaging over both quantum states and impurities in the grains. In principle, the grains can be clean and the electrons can scatter mainly on the surface of the grains. However, provided the shape of the grains corresponds to a classically chaotic motion of the electrons, the clean limit should be described in the 0D case by the same formulas.

We carry out the calculation of the conductivity, making expansion in both the fluctuation modes and the tunneling term H_T . The right part of Eq. (1) can provide a small parameter of the expansion in the tunneling. As in conventional superconductors, we can single out contributions due to corrections to the DOS and Aslamazov-Larkin (AL) σ_{AL} and Maki-Thompson (MT) σ_{MT} corrections. Diagrams describing these contributions are represented in Fig. 1. As a result, the total conductivity σ can be

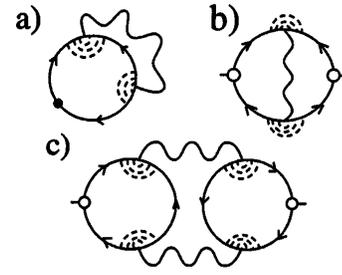


FIG. 1. Diagrams describing corrections to conductivity due to superconducting fluctuations. The wavy lines denote the propagator of the fluctuations, and the dashed lines describe impurity scattering.

written as

$$\sigma = \sigma_{\text{DOS}} + \sigma_{\text{AL}} + \sigma_{\text{MT}}, \quad (5)$$

where σ_{DOS} is given by equation

$$\sigma_{\text{DOS}} = \sigma_0 (4T)^{-1} \int_{-\infty}^{+\infty} [\nu(\varepsilon)/\nu_0]^2 \cosh^{-2}\left(\frac{\varepsilon}{2T}\right) d\varepsilon, \quad (6)$$

where $\sigma_0 = 4\pi e^2 d^{-1} (t/\delta)^2$ is the classical conductivity of the granular metal and $\nu(\varepsilon)$ is the density of states modified by the superconducting fluctuations and averaged over the impurities.

The main correction $\delta\nu(\varepsilon)$ to the DOS of the noninteracting electrons ν_0 is described by the diagram in Fig. 1a, while the terms σ_{MT} and σ_{AL} are given by Figs. 1b and 1c, respectively. The calculation of the diagrams can be performed for the Matsubara frequencies $\varepsilon_n = \pi T(2n + 1)$ using temperature Green functions. At the end one should, as usual [13], make the analytical continuation $i\varepsilon_n \rightarrow \varepsilon$. The magnetic field can be considered in the quasiclassical approximation, which results in additional phases in Green functions.

The diagrams in Fig. 1 contain essentially the averaged one-particle Green functions, the impurity vertices C , and the propagator of the superconducting fluctuations K . The functions C and K depend on the coordinates and time slower than the averaged one-particle Green functions. As a result, the magnetic field affects only the vertex C and the propagator K , whereas the phases of the Green functions drawn in Fig. 1 outside these blocks cancel. So, reading the diagrams in Fig. 1, one should replace the solid lines by the functions

$$G^{(0)}(i\varepsilon_n, \mathbf{p}) = [i\varepsilon_n - \xi(\mathbf{p}) + i(2\tau)^{-1} \text{sgn}\varepsilon_n]^{-1}. \quad (7)$$

The impurity vertex entering these diagrams is equal to $(2\pi\nu_0\tau)^{-1} C(i\varepsilon_n, i\Omega_k - i\varepsilon_n)$, where C is the so-called Cooperon. It obeys the following equation:

$$\begin{aligned} \{D_0[-i\nabla - (2e/c)\mathbf{A}]^2 + |2\varepsilon_n - \Omega_k|\} C(\mathbf{r}, \mathbf{r}') \\ = 2\pi\nu_0\delta(\mathbf{r} - \mathbf{r}'), \end{aligned} \quad (8)$$

where $D_0 = v_0^2\tau/3$ is the classical diffusion coefficient. The vector potential $\mathbf{A}(\mathbf{r})$ should be chosen in the London gauge. If the shape of the grain is close to spherical, the vector potential is expressed through the magnetic field \mathbf{H} as $\mathbf{A}(\mathbf{r}) = (\mathbf{H} \times \mathbf{r})/2$.

All relevant energies in the problem are assumed to be much smaller than the energy of the first harmonics $E_c = D_0\pi^2/R^2$ playing the role of the Thouless energy of a single grain, and this allows one to keep only the zero harmonics in the spectral expansion. One can find the eigenvalue $\mathcal{E}_0(H)$ of this harmonics using the first order of the standard perturbation theory

$$\mathcal{E}_0(H) = (2e/c)^2 D_0 \langle \mathbf{A}^2 \rangle_0, \quad (9)$$

where $\langle \dots \rangle_0$ stands for the averaging over the volume of the grain. For a grain of a nearly spherical form, one obtains

$$\mathcal{E}_0(H) = \frac{2}{5} \left(\frac{eHR}{c} \right)^2 D_0 = \frac{2}{5} \left(\frac{\phi}{\pi\phi_0} \right)^2 E_c, \quad (10)$$

where $\phi_0 = \pi c/e$ is the flux quantum and ϕ is the magnetic flux through the granule.

Within the zero-harmonic approximation, the function C does not depend on coordinates and equals

$$C(i\varepsilon_n, i\Omega_k - i\varepsilon_n) = 2\pi\nu_0 [2\varepsilon_n - \Omega_k + \mathcal{E}_0(H)]^{-1}. \quad (11)$$

The propagator of the superconducting fluctuations is calculated by summing, as usual, the superconducting ladder containing the products $G_{i\varepsilon_n} G_{i\Omega_k - i\varepsilon_n}$. Neglecting weak localization corrections, one reduces the averaging of the propagator K over the impurities to the averaging of these products that finally give C . The tunneling can be neglected here provided the right part of Eq. (1) is fulfilled. However, the tunneling from grain to grain should be taken into account when calculating K . This can be done by making an expansion in H_T [Eq. (3)]. Each new vertex arising in this expansion should also be dressed by impurity lines.

Although the final result can be written for arbitrary T and H , let us concentrate on the most interesting case $T \ll T_c$, $H > H_c$. Assuming for simplicity that the granules are packed into a cubic lattice and using the momentum representation with respect to coordinates of the grains, we obtain in this limit

$$K(i\Omega_k, \mathbf{q}) = -\nu_0^{-1} \left[\ln \left(\frac{\mathcal{E}_0(H)}{\Delta_0} \right) + \frac{|\Omega_k|}{\mathcal{E}_0(H)} + \eta(\mathbf{q}) \right]^{-1}, \quad (12)$$

$$\eta(\mathbf{q}) \equiv (8/3\pi) \sum_{i=1}^3 J[\delta/\mathcal{E}_0(H)] (1 - \cos q_i d),$$

where $J = (\pi^2/4)(t/\delta)^2$, \mathbf{q} is the quasimomentum, $d = 2R$, and $|\Omega_k| \ll \mathcal{E}_0(H)$. The pole of the propagator K [Eq. (12)] at $\mathbf{q} = 0$, $\Omega_k = 0$ determines the field H_c at

which the BCS gap disappears,

$$\mathcal{E}_0(H_c) = \Delta_0. \quad (13)$$

The result for H_c [Eqs. (10) and (13)] agrees with one obtained long ago by another method [14]. The term $\eta(\mathbf{q})$ in Eq. (12) is very important if H is close to H_c . For $\varepsilon_n > 0$ the contribution of the diagram, Fig. 1a, is

$$\delta\nu(i\varepsilon_n) = \frac{2iT}{\nu_0} \sum_{\Omega_k < \varepsilon_n} \times \int K(i\Omega_k, \mathbf{q}) C^2(i\varepsilon_n, i\Omega_k - i\varepsilon_n) \frac{d^3q}{(2\pi)^5}. \quad (14)$$

After calculation of the sum over Ω_k in Eq. (14), one should make the analytical continuation $i\varepsilon_n \rightarrow \varepsilon$. At low temperatures it is sufficient to find $\delta\nu \equiv \delta\nu(0)$.

Remarkably, Eqs. (12)–(14) do not contain explicitly the mean free time τ . This is a consequence of the zero-harmonic approximation, which is equivalent to using the random matrix theory (RMT) [11]. [The parameter τ enters only Eq. (10), giving the standard combination $\mathcal{E}_0(H)$ describing in RMT the crossover from the orthogonal to the unitary ensemble.] This justifies the claim that the results can also be used for clean grains with a shape providing a chaotic electron motion.

Using Eqs. (6) and (12)–(14), one can easily obtain an explicit expression for σ_{DOS} for $H - H_c \ll H_c$. In this limit, one expands the logarithm in the denominator of Eq. (12) and neglects the dependence of C on ε_n and Ω_k because the main contribution in the sum over Ω_k comes from $\Omega_k \sim \mathcal{E}_0(H) - \mathcal{E}_0(H_c) \ll \Delta_0$. The result for $\delta\sigma_{\text{DOS}} = \sigma_0 - \sigma_{\text{DOS}}$ can be written as

$$\frac{\delta\sigma_{\text{DOS}}}{\sigma_0} = -\frac{2\delta}{\Delta_0} \begin{cases} -\pi^{-1} \langle \ln \tilde{\eta}(\mathbf{q}) \rangle_q, & T/\Delta_0 \ll \tilde{\eta} \\ \frac{2T}{\Delta_0} \langle \tilde{\eta}^{-1}(\mathbf{q}) \rangle_q, & \tilde{\eta} \ll T/\Delta_0 \ll 1 \end{cases}, \quad (15)$$

$$\tilde{\eta}(\mathbf{q}) = \eta(\mathbf{q}) + 2h, \quad \langle \dots \rangle_q \equiv V \int (\dots) d\mathbf{q}/(2\pi)^3,$$

where $h = (H - H_c)/H_c$. We see that the correction to the conductivity is negative and its absolute value decreases when the magnetic field increases. The correction reaches its maximum at $H \rightarrow H_c$. At zero temperature, the maximum value of $\delta\sigma_{\text{DOS}}/\sigma_0$ is of order δ/Δ_0 . As temperature grows, the correction can become larger and reach, for $T \sim \Delta_0$, the order of magnitude of J^{-1} . Both of the values are smaller than unity because we work in the region specified by Eq. (1) and this justifies the diagrammatic expansion we use. The correction can become comparable with σ_0 when $J \sim 1$, which would mean that we are not far from the metal-insulator transition. For such values of J , one can use Eq. (15) only for rough estimates. Apparently, the parameters of the samples of Ref. [1] correspond to the region $J \sim 1$, $\delta/\Delta_0 \sim 1$.

In the opposite limit $H \gg H_c$ the correction to σ_0 can still be noticeable. In this case, we can neglect the dependence of the superconducting propagator [Eq. (12)] on Ω_k and on the tunneling term [because now $\ln(\mathcal{E}_0(H)/\mathcal{E}_0(H_c)) \gg 1$] and finally obtain

$$\delta\sigma_{\text{DOS}}/\sigma_0 = -(1/3)[\delta/\mathcal{E}_0(H)]\ln^{-1}[\mathcal{E}_0(H)/\Delta_0], \quad (16)$$

which means that in the region $H \gg H_c$ the correction to the conductivity decays essentially as $\delta\sigma_{\text{DOS}} \sim H^{-2}$.

Let us emphasize that the correction to the conductivity coming from the DOS remains finite in the limit $T \rightarrow 0$, thus indicating the existence of the virtual Cooper pairs even at $T = 0$.

In order to calculate the entire conductivity [Eq. (5)], we must investigate the AL and MT contributions (Figs. 1c and 1b). In conventional superconductors near T_c , these contributions are most important, leading to an increase of the conductivity.

In the granular materials the situation is much more interesting. It turns out that both of the AL and MT contributions *vanish* in the limit $T \rightarrow 0$ at all $H > H_c$ and, thus, the correction to the conductivity comes from the DOS only. Leaving the presentation of the details for a future publication [15], we list here only the final results.

The AL correction to the conductivity σ_{AL} corresponds to the tunneling of the virtual Cooper pairs and must be proportional to t^4 , in contrast to the one-electron tunneling determining σ_{DOS} . All calculations are done in the same approximation as when calculating σ_{DOS} . In the limit $T \ll T_c$, $H - H_c \ll H_c$, the result for σ_{AL} can be written as

$$\frac{\sigma_{\text{AL}}}{\sigma_0} = \frac{4\pi^2 t^2 T}{9\Delta_0^3} \sum_{i=1}^3 \langle A(\mathbf{q}) [\tilde{\eta}(\mathbf{q})]^{-3} \sin^2 q_i d \rangle_q, \quad (17)$$

where $A(\mathbf{q}) = 4\pi T [3\Delta_0 \tilde{\eta}(\mathbf{q})]^{-1}$ for $T \ll \Delta_0 \tilde{\eta}$ and $A(\mathbf{q}) = 1$ for $T \gg \Delta_0 \tilde{\eta}$.

As usual, the MT diagrams have both regular and anomalous parts. For the problem considered, they are of the same order of magnitude but have opposite signs. Moreover, at $T = 0$ they cancel each other. The final result for the MT contribution takes the form

$$\frac{\sigma_{\text{MT}}}{\sigma_0} = \frac{16\pi^2 T^2 \delta}{9\Delta_0^3} \sum_{i=1}^3 \langle B(\mathbf{q}) \cos q_i d \rangle_q, \quad (18)$$

where $B(\mathbf{q}) = -\pi^{-1} \ln \tilde{\eta}(\mathbf{q})$ for $T \ll \Delta_0 \tilde{\eta}$ and $B(\mathbf{q}) = 2T [\Delta_0 \tilde{\eta}(\mathbf{q})]^{-1}$ for $T \gg \Delta_0 \tilde{\eta}$.

The temperature and magnetic field dependences of σ_{AL} and σ_{MT} are rather complicated but they are definitely positive. The competition between these corrections and σ_{DOS} determines the sign of the magnetoresistance. We

see from Eqs. (17) and (18) that both the AL and MT contributions are proportional at low temperatures to T^2 . Therefore the σ_{DOS} in this limit is larger and the magnetoresistance is negative for all H_c . In contrast, at $T \sim T_c$ the AL and MT corrections can become close to H_c larger than σ_{DOS} , resulting in a positive magnetoresistance in this region. Far from H_c the magnetoresistance is negative again.

The present paper cannot help to answer the question of whether or not there is a phase transition at H_c because the perturbation theory does not work very close to H_c . Investigation of this problem can bring new understanding of the granular materials.

In conclusion, we have shown that superconducting fluctuations in granular metals at low temperature and high magnetic field lead to a suppression of the density of states. The well-known Aslamazov-Larkin and Maki-Thompson corrections vanish in the limit $T \rightarrow 0$ and all of this leads to the reduction of the conductivity.

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