Insulating state of granular superconductors in a strong-coupling regime

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(Dated: 12 July 2006)

We analyze the possibility of the formation of a magnetic-field-induced insulating state in a two-dimensional granular superconductor with relatively strong intergranular coupling and show that such a state appears in a model with spatial variations of the single-grain critical magnetic field. This model describes realistic granular samples with the dispersion in grain sizes and explains the mechanism leading to a giant peak in the magnetoresistance.

PACS numbers: 74.81.-g,74.78.-w,74.45.+c,74.20.-z

I. INTRODUCTION

Recent experiments on superconducting films revealed a giant peak in the low temperature magnetoresistance with the growth from several times (in polycrystalline samples) to several orders of magnitude (in amorphous films), as compared to the value near the superconductor-insulator transition, and the re-entrant drop upon further increase of magnetic field. The magnetic field-induced insulating phase and the related superconductor-insulator transition are long known, and extensive theoretical and experimental efforts resulted in a remarkable progress in understanding many features of these phenomena. Yet the observed giant peak in magnetoresistance is puzzling and poses a challenge for theory. Theoretical models suggested possible development of spatial inhomogeneities in amorphous samples and indicated the role they may play in formation of the resistivity maximum. Consequences of the existence of the superconductor islands network were explored by the numerical studies in Ref. 12; the computed behavior of magnetoresistance was in a qualitative agreement with the experiment.

In the present paper we propose and investigate a model for polycrystalline and/or granular films which offers a mechanism leading to the giant peak in the magnetoresistance. We model the superconducting film in a strong magnetic field as an array of alternating superconducting (S) and normal (N) granules and demonstrate that the superconducting insulator (SI) phase appears in the strong coupling regime. In real multi-disperse films the formation of a network of S and N grains occurs naturally as a consequence of the spatial variations of the single grain critical fields that inevitably appear due to dispersion of grains’ sizes. Magnetic field close to the average single-grain critical field drives about a half of the grains into a normal state, giving rise to the network of S and N grains. Note that the “standard” model of an array of identical S granules cannot describe the formation of an insulating state in the regime of a strong intergranular coupling. The charging energy of a single grain is renormalized down to the values \(E_c \sim \Delta/G\) where \(\Delta\) is the order parameter in a single grain and \(G\) is the intergranular tunneling conductance. The Josephson coupling \(E_J \sim G\Delta \gg E_c\) if \(G \gg 1\), and this results in the superconducting rather than the insulating state.

We consider the case where most of the grains are so small that the Zeeman effect is at least of the same order as the orbital one, and, as a result, the transition between the superconducting and normal states within a single grain is of the first order. Such samples are the good candidates for demonstrating the SI phase since the applied strong magnetic field \(H\) close to the Clogston field \(H_0\) will form a strongly inhomogeneous mixture of S and N grains with the order parameter in the S granules close to that in the absence of the magnetic field, \(\Delta_0\). Figure 2 shows the phase diagram of a single superconductor grain (the derivation is presented below) drawn in the coordinates Zeeman energy \(h = \mu_B H\), where \(\mu_B\) is the Bohr magneton, vs. the size-dependent orbital effect pairbreaking parameter \(\alpha\) [in spherical grains, \(\alpha = D(cH/2c)^2/5\), where \(c\) is the grain diameter and \(D\) is the bulk diffusion constant]. Varying the grain size one crosses the line of the phase transition which at strong enough fields is indeed always of the first order.

We now recall a well known result of the percolation theory that in the site problem for the two-dimensional black and white alternate square (or honeycomb) lattice there exists a range of the relative concentrations of the

FIG. 1: (a) Superconductor film consisting of grains of slightly different sizes. (b) The same granular array under applied magnetic field close to the average single-grain critical field. Varying magnetic field results in a change of the relative concentration of superconducting (white) and normal (gray) grains.
suggests the conductivity behavior

\[ G < G^* \]

where \( \delta \) is the mean energy level spacing in a single grain. Since the ratio \( \Delta_0/\delta \) is large (otherwise even the mean field approximation for a single grain cannot be used), \( G^* \) is larger than the critical value

\[ G_c \approx \frac{1}{4\pi} \ln \frac{E_c}{\delta} \]

that marks the onset of strong Coulomb correlations in the two-dimensional granular array in the absence of superconductivity\(^{19,20} \). Thus, for samples with intergranular conductance laying within the interval

\[ G_c < G < G^* \]

the insulating state is indeed induced by the local superconducting correlations.

## II. PHASE DIAGRAM OF A SUPERCONDUCTING GRAIN

Now we turn to the analytical derivation of our results. We begin with the discussion of a single granule of the size less than the coherence length. The difference between its thermodynamic potentials in superconducting and normal states can be written as

\[ \Omega = \frac{|\Delta|^2}{\lambda \delta} + \frac{\pi T}{\delta} \sum_\omega \left[ \alpha f f^\dagger - 2\tilde{\omega}(g - \text{sgn}\omega) - \Delta f^\dagger - \Delta^* f \right] \]

where \( \lambda \) is the electron-phonon coupling constant, \( \tilde{\omega} = \omega - \text{ih} \), \( \omega \) is the fermionic Matsubara frequency, and \( g, f, f^\dagger \) are the Usadel Green functions subject to the constraint \( g^2 + f f^\dagger = 1 \). Varying \( \Omega \) with respect to \( f^\dagger \) under the above constraint results in the zero-dimensional Usadel equation

\[ \alpha g f = \Delta g - \tilde{\omega} f, \]

while varying in \( \Delta^* \) yields the self-consistency equation

\[ \Delta = \lambda \pi T \sum_\omega f, \]

(below we choose \( \Delta \) real and \( f = f^\dagger \)). The second-order phase transition line can be obtained by solving Eq. \((7)\) in the first order with respect to \( \Delta \) and inserting the resulting \( f \) function into Eq. \((8)\). At zero temperature, \( T = 0 \), we arrive at

\[ \alpha^2 + h^2 = \frac{\Delta_0^2}{4} \]

The second-order phase transition line turns into the first-order one (see Fig. 2) that can be found by solving Eqs. \((7)\) and \((8)\) under the condition \( \Omega = 0 \). At \( \alpha = 0 \) we reproduce the Clogston result\(^{15} \) for the critical field

\[ h_0 = \frac{\Delta_0}{\sqrt{2}}. \]
The boundary between the gapful (S) and gapless (GS) phases is obtained from the condition that the gap in the electron spectrum $\Delta_g = 0$.\textsuperscript{16,23} $\Delta_g = \left( \Delta^{2/3} - \alpha^{2/3} \right)^{3/2} - h,$ \hfill (10)

where $\Delta$ has to be found from Eq. 3. The latter is not affected by the Zeeman term (since the spin susceptibility in this regime is zero at $T = 0$) in the gapful regime and thus assumes a simple form\textsuperscript{26}

$$\ln \frac{\Delta_0}{\Delta} = \frac{\pi \alpha}{4 \Delta}. \hfill (11)$$

We see that at small enough grain sizes (small $\alpha$) the gapless region does not exist. In what follows we will focus on the situation where the superconducting grains all have the gap. The validity of this assumption is supported by the fact that superconducting insulator phase was observed at very high magnetic fields of the order of the Clogston limit. The gapless region is not favorable for the formation of the SI state.

### III. ARRAY OF SUPERCONDUCTING GRAINS

We describe the array of S and N grains by the phase action\textsuperscript{24}

$$S = \sum_i \int d\tau \frac{\dot{\phi}_i^2(\tau)}{4E_c} + S_{ns}, \hfill (12a)$$

where the first term is the Coulomb part of action with the sum going over all grains. The term $S_{ns}$ describes the coupling of normal and superconducting grains:

$$S_{ns} = \frac{\pi G}{2} \sum_{ij} \int d\tau d\tau' \alpha(\tau - \tau') \left( 1 - e^{i \phi_{ij}(\tau) - i \phi_{ij}(\tau')} \right), \hfill (12b)$$

where summation goes over the nearest neighboring grains, the phase difference $\phi_{ij} \equiv \phi_{ij} (\tau) - \phi_{ij} (\tau)$ and the kernel

$$\alpha(\tau) = T^2 \sum_{\omega, \omega'} e^{-i(\omega - \omega')\tau} g_s(\omega) g_n(\omega') \hfill (13)$$

is expressed through the Usadel Green functions $g_s(\omega)$ and $g_n(\omega)$ of the S and N grains.

In the limit $G \gg G_c$ the term $S_{ns}$ can be simplified by expanding it up to the second order in $\phi_{ij}(\tau)$ and using the local approximation for the kernel $\alpha$ to

$$S_{ns} = \frac{G g'_s(0)}{4} \sum_{ij} \int d\tau \dot{\phi}_{ij}^2(\tau), \hfill (14)$$

where $g'_s(0)$ is the derivative of the function $g$ with respect to $\omega$ taken at $\omega = 0$. It can be found from Eq. 7, $g'_s(0) = (-\alpha + \Delta/f^3(0))^{-1}$, and in the limiting case that we assume, $h \approx h_0 \gg \alpha$, reduces to

$$g'_s(0) \approx \frac{2\sqrt{2}}{\Delta_0}. \hfill (15)$$

In the Fourier representation we obtain

$$S = \frac{e^2}{4} \int d\tau \int \frac{d^2q}{(2\pi)^2} \left[ E_c^{-1} + B(1 - E_q) \right] |\dot{\phi}_q|^2, \hfill (16)$$

where we have introduced the notations

$$E_q = \frac{1}{2} \sum_a \cos qa, \hfill (17)$$

with $a$ being the lattice vectors and

$$B = 8G g'_s(0). \hfill (18)$$

Integration over quasimomentum $q$ is over the first Brillouin zone. Equation (17) can be interpreted as the renormalization of the charging energy,

$$E_c \to \tilde{E}_c(q) = \frac{1}{E_c^{-1} + B(1 - E_q)}. \hfill (19)$$

The gap in the electron spectrum becomes

$$\Delta^{(e)} = \tilde{E}_{00}(q) = a^2 \int \frac{d^2q}{(2\pi)^2} \tilde{E}_c(q), \hfill (20)$$

where $\tilde{E}_{00}$ is a diagonal element of the Coulomb interaction matrix $E_{ij}$ that is given by the inverse Fourier transform of $\tilde{E}_c(q)$. Within the logarithmic accuracy

$$\Delta^{(e)} = \frac{1}{8\pi G g'_s(0)} \ln \frac{G E_c}{\Delta_0}, \hfill (21)$$

leading to Eq. 11 in the case of strong fields where Eq. 15 can be used. We see that $\Delta^{(e)}$ is indeed less than $\Delta_0$ as long as $G \gg G_c$. This justifies the local in time approximation used in obtaining Eq. 14. We note that in terms of the phase functional there is no difference between the S and N grains. However, one has to remember that the S grains have the “big” single particle gap $\Delta_g$. At small nonzero temperatures, the electron excitations “live” only in the N grains, above the gap $\Delta^{(e)}$. Despite the superconducting gap, electrons can tunnel between two N grains via a virtual state of an S grain, see diagram in Fig. 3(a) (this higher-order tunneling process is analyzed below). Therefore the transport has the activation form with the gap $\Delta^{(e)}$.

Now we consider the higher-order tunneling processes so far neglected and find the conditions, at which the contribution of the diagrams in Fig. 3 is not essential.

#### A. Stability of the superconducting insulator state with respect to the formation of the normal state

The diagram in Fig. 3(a) should be analyzed in order to check the stability of the SI state with respect to
formation of the normal state. Such tunneling processes lead to an effective coupling of N grains that due to condition $\Delta^{(e)} \ll \Delta_0$ can be viewed as an effective tunneling coupling with the conductance:

$$G_{ij}^{(n)} = \frac{t_{ij}}{4} G^2 g_s'(0) \delta,$$

where the dimensionless matrix element $t_{ij} = 2$ and $t_{ij} = 1$ for nearest and next-to-nearest neighbors in the N sublattice, respectively. This effective coupling results in the additional term in the phase action

$$S_{nn} = -\frac{1}{2\pi} \sum_{ij} G_{ij}^{(n)} \int d\tau d\tau' \frac{e^{i\phi_{ij}(\tau) - i\phi_{ij}(\tau')}}{\tau - \tau'}^2. \quad (23)$$

To find the correction to the Coulomb gap $\Delta^{(e)}$ in the first order in $S_{nn}$, we can consider the normal sublattice of the array only. The problem then becomes equivalent to the calculation of the correction to the Mott gap in the array of N grains. Using the result of Ref. 21 we obtain the correction to $\Delta^{(e)}$:

$$\delta \Delta^{(e)} = -\frac{4h_2}{\pi} \sum_i G_{0i}^{(n)} \langle \tilde{E}_{00} - \tilde{E}_{0i} \rangle. \quad (24)$$

Using that $E_s G \gg \Delta_0$ from Eqs. 19 and 21 we obtain

$$\delta \Delta^{(e)} = -\frac{2h_2}{\pi} G \delta. \quad (25)$$

Comparing Eqs. 1 and 26, we conclude that $\delta \Delta^{(e)} \ll \Delta^{(e)}$ if

$$G \ll \left( \frac{\Delta_0}{\delta} \right)^{1/2}. \quad (26)$$

We note that the effective coupling also defines the prefactor in the conductivity behavior:

$$\sigma = \frac{4e^2}{h} G^2 \delta g_s'(0) \exp \left( - \frac{\Delta^{(e)}}{T} \right), \quad (27)$$

where the function $g_s'(0)$ was defined in Eq. 15.

B. Stability of the superconducting insulator state with respect to the formation of the superconductor state

Now we analyze the stability of the insulating state with respect to formation of superconductivity that may be established due to the Josephson coupling of the S grains, see the diagram in Fig. 3(b). To this end we introduce an effective Hamiltonian acting on Cooper pairs and defined on the sublattice of the S grains

$$\hat{H} = 4 \sum_{ij} n_i \hat{E}_{ij} \hat{n}_j - \frac{J}{2} \sum_{ij} t_{ij} e^{i\phi_i - i\phi_j}. \quad (28)$$

The Hamiltonian is written in terms of the superconducting phase $\varphi$, which is related to the “normal” phase $\phi$ as $\varphi = 2\phi$. Here $i, j$ belong to the S sublattice, $\hat{n} = -i\partial / \partial \varphi$ is the Cooper pair density operator, $J$ is the effective Josephson coupling of S grains via a N grain, and $t_{ij} = 2$ and $t_{ij} = 1$ for nearest and next-to-nearest neighbors in the S sublattice, respectively. The insulating state of the model (28) is characterized by the energy gap in the spectrum of Cooper pair excitations due to the doubled Cooper pair charge in the limit $J = 0$ this gap is

$$\Delta^{(s)} = 4\Delta^{(e)}. \quad (29)$$

The Josephson coupling $J$ leads to the suppression of $\Delta^{(e)}$ that in the limit of weak coupling can be found via the straightforward perturbation theory:

$$\delta \Delta^{(s)} = -\frac{J}{2} \sum_i t_{0i} = -6J. \quad (30)$$

The condition $\delta \Delta^{(s)} \ll \Delta^{(e)}$ is satisfied if $J \ll \Delta^{(e)} \sim \Delta_0/G$. The diagram in Fig. 3(b) in the case of the large Zeeman energy, $h \approx h_0 \gg \alpha$, gives:

$$J = \frac{G^2 \delta}{4} \ln \left( \frac{\Delta_0^2}{4h} - 1 \right), \quad (31)$$

and we arrive at the estimate for the upper boundary for the conductance $4G^{25}$ One can verify that the two-electron (Andreev) tunneling processes between the normal grains via an intermediate superconducting one are also suppressed under the same condition.

IV. COMPARISON WITH EXPERIMENT

Now we discuss the relation between our results and experimental data of Refs. 1 and 2. A note in order is that the proposed model of a granular superconductor in the vicinity of a superconductor-insulator transition as a normal/superconductor granules alternating chessboard seems to capture adequately general features of the systems of real morphology and, therefore, allows for the comparison with the experimental data within its
Thus, the estimate of the gap $\Delta(G)$ taking the metallic domain, were close to the critical resistance at high magnetic fields, while falling into the superconducting state was observed in the samples where the values of films the formation of the “superconducting insulator” region. For example, amorphous samples can be not uniformly disordered but rather consisting of separate regions (due to fluctuations of chemical composition or impurity concentration) favoring superconductivity. In this case the system reminds the granular samples studied in the present paper.

Next, our theory is developed for granular systems and therefore should not apply to homogeneously amorphous samples. However, it is possible that realistic samples do not exactly correspond to one of the two limiting cases. For example, amorphous samples can be not uniformly disordered but rather consisting of separate regions (due to fluctuations of chemical composition or impurity concentration) favoring superconductivity. This case the system reminds the granular samples studied in the present paper.

Keeping this possibility in mind, we make estimates for the experiment of Ref. 2, where amorphous InO samples were investigated. We note that in the case of InO films the formation of the “superconducting insulator” state was observed in the samples where the values of the resistance at high magnetic fields, while falling into the metallic domain, were close to the critical resistance that separates insulating and metallic phases at $T \rightarrow 0$. Thus, the estimate of the gap $\Delta(e)$ in Eq. 10 requires taking $G \approx G_c$. Plugging this into Eq. 11, we obtain the gap

$$\Delta(e) = \frac{\Delta_0}{4\sqrt{2}} \frac{\ln(GE_0/\Delta_0)\ln(E_0/\delta)}{\ln(E_0/\delta)}.$$

Within the logarithmic accuracy, the ratio of the logarithms is about unity. Finally, in order to compare our estimate with the experimental findings, we express the superconducting gap $\Delta_0$ in the absence of the magnetic field through the critical temperature $T_c$. Using the standard relation $\Delta_0 = 1.76 T_c$, we arrive at the estimate

$$\Delta(e) = \frac{T_c}{3.2}$$

or, equivalently, $T_c/\Delta(e) \approx 3$, which certainly is in a reasonable agreement with the experiment. Indeed, according to Fig. 4 of Ref. 2, the ratio $T_c/\Delta(e)$ depending on the sample varies between 0.6 and 3. Bearing in mind that our model was strictly speaking designed for granular samples, one can hardly expect a better agreement.

V. DISCUSSION AND CONCLUSIONS

Here we comment on the conductivity behavior in the vicinity of the magnetization peak: At lower magnetic fields the concentration of the superconducting component increases resulting in the formation of the direct Josephson coupling between the superconducting grains. The Cooper pair gap $\Delta(e)$ decreases and eventually becomes smaller than the electron gap $\Delta(e)$. At the same time there is no reason for the strong suppression of the electron gap due to the Josephson coupling. Thus, closer to the insulator to superconductor (IS) transition at finite but low temperatures, the transport is mediated by the activation of Cooper pairs rather than by the single electron excitations, hence $\sigma \sim \exp \left(-\frac{\Delta(e)}{T}\right)$ at $T \ll \Delta(e)$. Thus, we expect that the resistivity within the IS transition may be described within the effective Bose model as in Ref. 2. At the opposite side of the magnetoresistance peak (i.e., at higher fields) the fraction of the normal component increases leading to the direct coupling of normal grains. This results in the decrease of the electron gap $\Delta(e)$ and eventually in the destruction of the insulating state. Note that for the SI state which we studied the spatial orientation of the applied magnetic field was not essential, however closer to the IS transition one expects it to become important.

In conclusion, we have investigated formation of an insulating state (corresponding to a giant peak in the magnetoresistance) in a two-dimensional granular array with alternating superconducting and normal granules. We have demonstrated that this model is a good representation for realistic granular (or polycrystalline) systems with the grain size dispersion in a strong magnetic field.

Acknowledgments

We thank M. V. Feigel’man for bringing our attention to this problem, and T. I. Baturina, K. B. Efetov, and V. F. Gantmakher for useful discussions. This work was supported by the U. S. Department of Energy, Office of Science via the contract No. W-31-109-ENG-38. Ya.V.F. was also supported by the RFBR grant No. 04-02-16348, the RF Presidential Grant No. MK-3811.2005.2, the Russian Science Support Foundation, the Dynasty Foundation, the Russian Ministry of Industry, Science and Technology, the program “Quantum Macrophysics” of the RAS, CRDF, and the Russian Ministry of Education.

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27 The Coulomb interaction on the normal grain can be neglected for \( h \gg E_c \sim \Delta_0 / G \). Note, that in the opposite limit of strong Coulomb interaction, \( \Delta_0 \gg E_c \gg h \), the result is \( J = (G^2 \delta / 2) \ln(\Delta / E_c) \).

28 For \( h = h_0 \equiv \Delta_0 / \sqrt{2} \), Eq. 31 gives zero and therefore the result for the Josephson current should be written more accurately. For small \( \alpha \) and \( h_1 \equiv h_0 - h \ll h_0 \) we obtain \( J = G^2 \delta \sqrt{2}(h_1 / \Delta_0 - 2 \alpha / 3 \Delta_0) \).