Destruction of the Meissner Effect in Granular High-Temperature Superconductors

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We discuss a state of an orbital glass which may exist in granular superconductors. The orbital glass (new topological glass) is related to the existence of the special loops of Josephson junctions with positive and negative Josephson couplings. With an odd number of negative couplings on the loop, a spontaneous orbital moment is created. Similar to anyon superconductivity, the new state with orbital moments is characterized by broken fundamental symmetries (C and P). There the orbital paramagnetism coexists with the superconductivity and at small magnetic field and low temperatures the Meissner effect may disappear.

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The orbital glass is a state that is in some sense analogous to a spin glass, but instead of spins there are "orbital moments." Exactly speaking these orbital moments are associated with circular currents that flow around loops of Josephson junctions in the superconducting state. The supercurrents generate flux tubes of magnetic field, which are normal-state regions within the superconductor. The susceptibility of the state with such flux tubes may be paramagnetic even if the sample is in the superconducting state.

Granular high-temperature superconductors (HTSC), having a complicated microstructure because of the anisotropy of the grains, are more favorable for the existence of the orbital glass. The flux tubes in the granular HTSC will penetrate the grains along a-b planes. The currents will flow over loops that cover one or several grains.

A three-dimensional array of the Josephson junctions is a good model for the granular HTSC. In a superconducting state a Josephson coupling J is created between grains. If there is an impurity state between two grains, which may be occupied by one electron with spin up (down), then the Josephson coupling changes sign $-J$. In the tunneling process of the superconducting pair over the impurity state there appears one permutation of two fermions, which gives the negative sign. Thus, a fundamental reason for the appearance of the negative Josephson coupling is the statistics of fermions [1-3].

The appearance of these negative Josephson couplings may give rise to creation of spontaneous circular currents. To create the current it is necessary to have a loop with an odd number of negative Josephson couplings [4]. The current will give rise to a spontaneous magnetic flux, creating a flux phase state on the ring [5].

Considering a single ring, let us estimate the current and the magnetic flux. The Hamiltonian for a ring, consisting of N Josephson junctions, has a form that is a simple generalization of [6,7]:

$$H=\sum_{i,j}^{N}\frac{\alpha_{i,j}}{2}n_i n_j-\sum_{\langle ij\rangle}^{N}J_{i,j}\cos(\theta_i-\theta_j-A_{ij}) , \quad (1)$$

where the summation is over all Josephson junctions of the loop. The first term of the Hamiltonian represents a Coulomb energy. For simplicity, we consider only on-site interaction with $\alpha=e^2/C$, where C is the capacitance of superconducting islands or grains, which is proportional to the size of grains. The second term represents a Josephson coupling between next-neighbor superconducting islands. We consider the cases when the constants $J_{i,j}$ may have both positive and negative values $J_{i,j}=\pm J$. n_i and θ_j are canonically conjugate variables $[n_i, \exp(\theta_j)]=\delta_{ij}\exp(\theta_j)$ [8], N is the number of islands, and A_{ij} is proportional to the line integral of the vector potential $\mathbf{A}$ between the ith and the jth sites. Here

$$A_{ij}=\frac{2e}{hc}\int_i^j \mathbf{A}\cdot dl , \quad (2)$$

where the total sum of the A_{ij} along the ring is a constant, $\sum_r A_{ij}=2\pi f$, which is proportional to the magnetic flux f piercing the ring in units of magnetic flux quantum Φ_0, $2\pi f=2\pi H\pi R^2/\Phi_0$ where R is the radius of the ring and H is a magnetic field.

The ground state is described by the following link-chain variational wave function:

$$\Psi(\theta_1,\ldots,\theta_N)=\frac{C_N}{(2\pi)^{N/2}}\prod_{\langle i,j\rangle}^{N}L_{i,j} , \quad (3)$$

where $N\geq 3$ and C_N is a normalization factor. The link function $L_{i,j}$ between two neighbor islands $\langle i,j\rangle$ has the form

$$L_{i,j}=[\exp(i\theta_i)+\delta\exp(i\theta_j)]\exp[i(-\theta_i-\theta_j)/2] , \quad (4)$$

where we assume the boundary condition $\theta_{N+1}=\theta_1$. The factor δ plays the role of variational parameter. The number of the link functions $L_{i,i+1}$ is equal to a number of Josephson junctions. The total many-body wave function, which is a product of the link functions, has the period 2π, as is necessary from gauge invariance. The function (4) is a two-plane-wave approximation of the exact solution for $N=2$.

The expectation value of the Hamiltonian (1) can be expressed through off-diagonal one-particle and twoparticles density matrices. In our variational approach these functions can be calculated analytically for an arbitrary N. We assume that M of the Josephson junctions is associated with a negative sign of J (π contacts). After some algebra with functions (3) and (4) for the ground-state energy per grain, we have

$$E(f)=\alpha\frac{k^2}{1+2k^N}-2J\cos\left(\frac{2\pi f\pm\pi M}{N}\right)\frac{2k+2k^{N-1}}{1+2k^N}, \tag{5}$$

where $-\frac{1}{2}\leq f\leq 0$ and $k=\delta/(1+\delta^2)$. We also used here an additional gauge transformation [9].

In the limit $N\rightarrow\infty$ the system transforms into a linear array of Josephson junctions. Bradley and Doniach have shown that the linear array has a Beresinskii-Kosterlitz-Thouless (BKT) phase transition [10] between the charge-charge coherent state and the superconducting phase-phase coherent state [6]. With the variational wave function we reproduce their result. That cannot be done with the function used in Ref. [11]. One sees that Eq. (5) has two different types of minima. One of them is marginal and occurs at small charging energy. The wave function of this state does not depend on α. This minimum corresponds to the superconducting phase-phase long-range coherent state. The other state is associated with the Coulomb blockade. Both states for a linear array of Josephson junctions have been described in Ref. [6]. For a ring of finite size there is no real phase transition but there is a transition between two quantum states. The point of the transition is smeared because of the finiteness of the ring size [12]. Therefore, the mean-field-like variational function works well.

The system may have a real BKT transition [10] if there is dissipation on the ring. This case is better described by the renormalization-group technique. This can be done by adding dissipation and by mapping the system into a vector Coulomb gas. We will not discuss here the BKT transition between these two states on the dissipating ring. We study a different problem, namely, the role of π contacts.

If the number of π contacts is an even number, then the magnetic properties are the same as those for the ring without π contacts [11]. If the number of π contacts is odd, the dependence of the ground-state energy upon the external magnetic flux has cuspidal maxima at integer flux quanta. In other words the dependence $E(f)$ at small $2\pi f/N$ is a linear function of f. This is crucial for a creation of spontaneous magnetic flux and an orbital current.

From Eq. (5) we get the ground-state energy for a phase-phase coherent state:

$$E(f)=\alpha d_1-J\cos\left(\frac{2\pi f+\pi}{N}\right)d_2,\quad -\tfrac{1}{2}<f<0, \tag{6}$$

where d_1 and d_2 are numerical coefficients, $d_1=1/(4+2^{3-N})$ and $d_2=(1+2^{2-N})/(1+2^{1-N})$. In the case $2\pi f/N\ll 1$, from Eq. (6) we obtain

$$E_{\rm gr}^{e}=A_1+B_1 f, \tag{7}$$

where $A_1=\alpha-J\cos(\pi/N)$ and $B_1=2\pi J/N\sin(\pi/N)d_2$.

This linear dependence of the ground-state energy on the flux f for the ring with an odd number of π contacts allows creation of the spontaneous flux. The flux can be determined by a minimization of the total energy E_Σ, consisting of the supercurrent energy $E_{\rm gr}^{e}$ and the magnetic field energy $E_M=C_0 f^2$:

$$E_\Sigma=C_0 f^2+A_1+B_1 f, \tag{8}$$

where C_0 is a constant, characterizing the inductance of the ring. The inductance may have a more complicated form [13], but we should stress that the magnetic energy is always proportional to f^2. Therefore, the dependence (8) on the flux f has the general form. From Eq. (8) we find the value of the spontaneously generated magnetic flux,

$$f_{\rm sp}=-B_1/2C_0 \tag{9}$$

with energy $E_{\Sigma\rm sp}=E_{\rm free}+\Delta E$, where $E_{\rm free}$ is the energy of Josephson junctions without magnetic field: $E_{\rm free}=A_1$ and ΔE is the decrease of the energy due to the creation of the spontaneous flux $f_{\rm sp}$, $\Delta E=-B_1^2/(4C_0)$. One sees that the last contribution is always negative, i.e., in the system a spontaneous orbital moment is always created. The same effect takes place for the other state, of course, if the ring has an odd number of π contacts. Because of the linear dependence of the ground-state energy upon the magnetic flux f a spontaneous magnetic field and a spontaneous orbital current arise. But instead of the magnitudes A_1 and B_1 in (7)-(9) we have here $A_2=4J^2/\alpha$ and $B_2=(J^2/\alpha)(8\pi/N)\sin(2\pi/N)$.

On the ring with $N=1$ and $N=2$ the orbital moment is not created at all. Quantum fluctuations destroy it. On the other hand, the value of the orbital moment decreases with N. The small temperature T of the order $\Delta E\sim N^{-2}$ (cf. [13-15]) destroys it. If $J\sim 10$ meV and $N\sim 3$, then for the temperature T we get $T\sim J/N^2\sim 15\,^\circ$C. Thus, the orbital moments are destroyed with temperature before the superconductivity disappears. The presence of disorder may dump the orbital current. Therefore the effect found in the paper may disappear with increasing disorder. The criterion is derived from comparison of the Thouless energy with ΔE.

If on a two-dimensional square array of Josephson junctions there exists a single π contact, then on two loops adjacent to the contact orbital currents will be created. The currents may have positive or negative circulation. The circular current generates a magnetic flux. The magnetic field generated will be located around the π contact. The total flux piercing the right plaquette will be about $+\pi$, corresponding to a vortex. The flux through the left plaquette will be about $-\pi$, corresponding to an antivortex. Therefore, one may conclude that the single π contact generates a vortex-antivortex pair.

Let the single π contact be located in the volume of the granular HTSC. Hence, it is obvious that around the π contact a normal state associated with a circular flux tube will be created. The volume of the normal state may be roughly estimated with the aid of energetic relations. The energy reduction due to the creation of spontaneous magnetic flux ΔE must be equal to the condensation energy associated with the destruction of the superconducting state and the creation of the normal state. Generally speaking, the existence of the π contacts does not always give rise to spontaneous orbital currents. Moreover there are many configurations with π contacts where there are no currents at all.

The loops of flux tubes will have different radii since the number of π rings associated with one flux tube may be arbitrary. With increasing magnetic field the loops begin to break. As a loop breaks a pair, consisting of single vortex and antivortex, is created. It is possible that the loop will split into two loops with smaller radii or it will split into another loop and a single vortex. Let us estimate the contribution to the susceptibility due to the π ring, which creates a decoupled vortex or an antivortex. For the phase-phase coherent state on the ring at small magnetic fields we get from Eq. (6) that the susceptibility χ equals

$$\chi = \frac{j(f)}{H} = -C\left(\frac{1}{4\pi} - \frac{M_{\text{or}}}{H}\right), \tag{10}$$

where $j(f) = -\partial E(f)/\partial f$ is an orbital current, $M_{\text{or}} = N\tan(\pi l/N)/(2\pi S)$ is an orbital moment induced by the π ring, $C = 8\pi^2 J d_2 S\cos(\pi l/N)/N$ is some constant, S is the area of an average Josephson loop, and the number l is equal to 0 or 1 for even and odd numbers of π contacts, respectively. In comparison with the case $l=0$ at an odd number of π contacts there is an additional positive contribution, which is proportional to $1/H$. We see that the presence of such a loop destroys the diamagnetic screening. Thus, at small field H the susceptibility $\chi \sim 1/H$ and the system behaves as a half flux quantum penetrating the ring. In other words, at small fields instead of varying like f/H for the ring without π contacts, the susceptibility varies like $1/H$.

Moreover, for a small magnetic field the character of the susceptibility is positive paramagnetic and the Meissner effect disappears. One can easily find the critical field H_{c0}, at which this transition to an orbital paramagnetic state occurs. Setting $\chi=0$ in Eq. (10) for $N \gg 1$ we immediately obtain

$$H_{c0} = \Phi_0/2S\,, \tag{11}$$

where Φ_0 is an elementary flux quantum. Thus, the critical field of the transition to the state of orbital glass is inversely proportional to the area of the average loop of Josephson junctions of granular HTSC. Although the field H_{c0} is always of the order of the inverse area S, the reduction energy ΔE strongly decreases with the charging energy. Therefore, the region of temperature ($T < \Delta E$) where the effect exists also decreases with charging energy.

Therefore a system with π rings behaves as a magnetic one with local orbital magnetic moments. The values of these orbital moments are very small, because $f_{\text{sp}} \sim 1/N^2$. Thus, even a small magnetic field can polarize this system, inducing different orbital moment flip processes. For granular HTSC with orbital currents in a magnetic field there exists a phase transition from a state with chaotic (randomly distributed with an arbitrary value) magnetic orbital moments (*orbital glass*) to a state of orbital ferromagnetism. Both states coexist with the superconducting state. In this system, as for ferromagnetism, different hysteresis cycles should exist.

In summary, we have studied the properties of granular HTSC, based on a model which suggests the existence of superconducting loops with weak links and with π contacts. There are two effects: The first effect is the Coulomb blockade, which can be both induced and removed by magnetic fields. The other effect is a generation of spontaneous magnetic flux on Josephson loops at an odd number of π contacts (on π rings). The circular currents over these Josephson loops create magnetic moments. If there exists dissipation in weak links, the value of the orbital currents decreases or may even increase. This depends on the local field from other loops, which can both increase and decrease. As a result we offer an *orbital glass*, where magnetism coexists with superconductivity and the Meissner effect is absent. This state is characterized by broken fundamental symmetries, like C and P, and has long-time tails in the dynamics associated with a broad spectrum of relaxation rates. Therefore, even for a small number of Josephson loops, which can exist around planar defects even in a single crystal of HTSC [16–19], there is no way to distinguish between anyon superconductivity (see [9], and references therein) and other ones. With an increase of the magnetic field the Meissner effect may again reappear.

After the submission of this work we became aware that there exist many experimental results about the complicated behavior of granular HTSC at small fields (a remanent magnetization [20] and a destruction of Meissner effect at low field and low temperatures [21–24]), which are in line with the proposed state of the orbital glass. First, an "anti-Meissner" effect has been observed in the granular Bi-Sr-Ca-Cu-O HTSC [21]. The authors of [21] have clearly concluded that for the explanation of the effect a new mechanism is needed. An estimation of H_{c0}, made in the framework of our model, gives $H_{c0} \sim 2$ G. This is approximately the observed value of H_{c0}. Thus we offer a mechanism for the anti-Meissner effect. The other observation of a disappearance of the usual Meissner effect in small magnetic fields in granular HTSC has been reported very recently [22–24].

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[12] The similar problem of the Coulomb blockade in the limits $E_J \gg E_C$ and $E_C \gg E_J$ for a single junction has been discussed in Ref. [13], where E_C and E_J are charging and Josephson energies, respectively. There are two different regimes of the behavior. They correspond to classical supercurrent behavior and to the Coulomb blockade. A supercurrent $I = I_c \cos\phi$ may flow if the phase ϕ does not fluctuate. The fluctuations of the phase ϕ may be estimated from the uncertainty relation [8]: $\delta n \delta\phi \geq e$. The absence of superconductivity (Coulomb blockade) may be easily understood in the limit $E_C \gg E_J$. In this case the charge is fixed $\delta n \to 0$ and therefore there are strong quantum fluctuations in the phase ϕ. Therefore, in an average over these fluctuations, the current equals zero: $\langle I(\phi)\rangle = 0$ (the Coulomb blockade). In the opposite limit $E_J \gg E_C$ the fluctuation of the phase ϕ is controlled by the value $\exp[-(8E_J/E_C)^{1/2}]$ [13]. A characteristic time τ for the spreading of the phase is proportional to the exponent of the ratio $(E_J/E_C)^{1/2}$. The time τ increases with the ratio, exponentially. This means that the phase ϕ will be localized during the time τ in one of the minima of the periodical potential and a supercurrent will flow. It is clear that the transformation of one regime into the other one is very smooth.

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