Weak Magnetic Order in High-$T_c$ Superconductors Produced by Spontaneous Josephson Currents

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Abstract – We develop a model for high-$T_c$ superconductors based on an electronic phase separation where low and high density domains are formed. At low temperatures this system may act as a granular superconductor forming an array of Josephson junctions. Cuprates are also known to have low superfluid densities and strong correlation effects. Both characteristics activate a negative Josephson coupling due to frustration that leads to spontaneous currents responsible for the weak ferromagnetic order. This original approach reproduces the observed onset of spontaneous magnetic signal and its dependence on the doping level.

Introduction. – Understanding of the normal state "pseudogap" phase of high-temperature superconductors remains one of the major puzzles of condensed matter physics. The reason abides in unusual properties and partial gapping of the low energy density of states below a certain temperature $T^*$ [1-3], taken as the boundary of the pseudogap phase. The lack of an accepted theory hints the nanoscale complexity and the intrinsically inhomogeneous electronic structures [4, 5], which might vary according to the specific family of cuprates.

Among the many anomalies of the pseudogap phase [1-3], the weak ferromagnetic signal on YBa$_2$Cu$_3$O$_{6+x}$ by zero-field muon spin relaxation [6] and by polar Kerr effect (PKE) requires an explanation. Both measure a hole concentration ($p$) dependent signal that starts jointly with $T^*(p)$ in agreement with many different experiments [1-3]. The values of $T^*(p)$ are well above $T_c(p)$ in the underdoped region and drops rapidly with increasing $p$, becoming comparable with $T_c$ near the optimally doped concentration $p = 0.16$.

In this letter we address this long standing problem by an entirely original approach. We assume an electronic phase separation with the charges segregated in domains (grains) of low and high densities separated by a potential barrier [8-10]. This charge segregation reduces the kinetic energies enhancing the possibility of superconducting amplitude formation in the isolated grains as a granular superconductor. This local superconductivity in domains is possibly similar to the case in which Bi-clusters of 2.5 to 40 nm becoming superconducting, in contrast with the non superconducting bulk Bi [11]. The free energy barriers between the domains produces charge confinement, as in the Bi-cluster case, and acts as weak links. We model this system as an array of Josephson junctions. Low local charge densities yield strong phase fluctuations [12] that combined with strong correlation effects gives rise to frustration [13]. Our breaking new idea proposes that these frustration effects are the origin of the weak ferromagnetic signal in YBCO [6, 7], which is a generalization to single crystals of the method used to explain the paramagnetic response of granular cuprates [14].

Many reviews [4,5] have discussed the existence of electronic inhomogeneities by different experiments on several samples. The disorder of the charge in cuprates can be treated by the Cahn-Hilliard differential equation that describes the formation of patches or grains. The charges trapped in these grains lose kinetic energy, form bound states and Cooper pairs, give origin to the pseudogap and superconducting phases [9,10]. With this, we illustrate a typical Ginzburg-Landau free energy simulation [8] on a $105 \times 105$ square lattice in Fig. 1.
tors showed anomalous magnetic properties attributed to frustration due to properties of the Josephson junctions [25]. In this case, the frustration effects arise due to the crystal’s axis mis-orientation of the grains with superconducting d-wave order parameter; this gives a negative contribution to the Josephson-junction energy. The current is negative with a phase shift $\pi$-a $\pi$ junction. Sigrist and Rice [14] demonstrated that a loop of current with an odd number of $\pi$ junctions is frustrated, i.e., there is no way to minimize the free energy of all junctions in the loop and a spontaneous current may arise if the coupling is sufficiently strong.

However, we cannot talk of mis-alignment of the crystal axis between different grains on a single crystal. In such system, negative Josephson coupling is possible due to the large fluctuation in the local density and correlation effects. This was demonstrated in detail by Spivak and Kivelson [21] using perturbation theory up to 4th-order in a model where correlations effects produce a negative Josephson coupling across a junction. For completeness we illustrate one of the situations described in their paper where a Cooper pair tunnels through a barrier which contains a spin up as shown in the Fig. 2.

It is important to remark that the physics of d-wave Josephson junctions presents many different and interesting aspects, depending on the type of superconductors, order parameter symmetry and geometry of the interface [23]. The d-wave pairing order parameter is very sensitive to inhomogeneities and interfaces. Quasiparticle scattering at interface distorts the order parameter and influences the Josephson effect as well as the quasiparticle tunneling current [24]. This phenomenon can generate midgap states, i.e., surface or interface states with zero energy relative to the Fermi energy [25] or Andreev zero-energy. The midgap states enhance the Josephson current near $T = 0$ and are observed through the zero bias conductance peak [25–27]. These states are robust phenomena and represent a crossover from a traditional Josephson junction (0-junction) to a $\pi$-junction, or a phase with broken time reversal symmetry.

On the other hand, the very high temperatures up to 200K of the signal measured by Xia et. al. [7] shows that its origin is not likely to be due to these interface states. Furthermore, the intrinsic inhomogeneous nanometer domains have small and randomly oriented interface, in opposition to those specially prepared junctions [25–27], and consequently it is difficult to generate midgap states. Thus, here we explore the idea that the origin of the $\pi$-junctions and the spontaneous magnetization in YBCO is due the very large phase oscillations due to very small local density of Cooper pairs [12].

**Calculations.** – The Cooper pairs tunneling through a localized state in the junction between two grains exchange order; this generates the negative sign -a characteristic of a $\pi$ junction [13]. Since the electronic phase separation with the granular structure provokes strong charge fluctuations, -this- and not the mis-orientation of the grains, is the mechanism originating $\pi$ junctions throughout the system.

Following the procedure of Sigrist and Rice [14], the properties of the whole loop of an odd number of $\pi$ junctions is given at one weak link. Assuming that the current $I$, which flows in the loop, is small compared to the critical current of the grains, we can write [14]

$$F(I, \Delta \phi) = \frac{L I^2}{2c^2} - \Phi_0 I_c \cos(\Delta \phi + \alpha),$$  \hspace{1cm} (1)

where $\Delta \phi$ is the phase shift across the junction, $L$ denotes the self-inductance of the loop, $I_c$ the critical current through the junction, and $\Phi_0$ is the flux quantum $hc/2e$.

The additional phase shift $\alpha$ is $\pi$ or 0 whether the loop
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is frustrated or not. Assuming also that the current flows through one point, this leads to

$$\Delta \phi = 2\pi n - 2\pi \frac{\Phi}{\Phi_0} = 2\pi n - \frac{2\pi}{\Phi_0} \left( \Phi_{ex} - \frac{LI}{c} \right),$$

(2)

where $n$ is an integer, $\Phi$ is the total flux threading the loop with three types of contributions; the external field, the flux from other loops called $\Phi_{ex}$ and the current $I$. With this $\Delta \phi$ Eq.[1] becomes

$$F(I, \Phi_{ex}) = \frac{LI^2}{2e^2} - \frac{\Phi_0 I_c}{2\pi c} \cos \left[ \frac{2\pi}{\Phi_0} \left( \Phi_{ex} + \frac{LI}{c} \right) + \alpha \right].$$

(3)

By minimizing $F$ with respect to $I$ for a given $\Phi_{ex}$, we relate $I$ and $\Phi_{ex}$. At zero external field we are interested in the case where $\Phi_{ex} \approx 0$ because the contributions from the other loops should be small. Then, for $\alpha = \pi$, a spontaneous flows around the loop if $\gamma \equiv 2\pi LI/c \Phi_0 c$ is larger than 1 (Fig.3). This spontaneous current -due to the loop's frustration- generates the observed weak magnetic signal.

Fig. 3: The induced current in a frustrated loop for three values of the parameter $\gamma$ as function of the external field. For the zero field case ($\Phi = 0$), it shows that for $\gamma > 1$ a spontaneous current may arise while there is not any for $\gamma < 1$.

Thus, as the parameter $\gamma$ surpasses unity, a spontaneous current flows around frustrated loops and local magnetic signals arise. The first spontaneous currents generate a magnetic field that influences the direction of other currents and small local inhomogeneous ferromagnetic signals arise throughout the sample, similar to the zero field susceptibility of granular superconductors near $T_c$ [15] ($T^*$ in our case). Muon spin relaxation ($\mu$-SR) [3] and polar Kerr-effect [7] measured such magnetic signal on YBCO compounds.

To compare our proposal with these experimental results we need to study the temperature and doping dependence of the parameter $\gamma$, since it controls the spontaneous currents. $\gamma$ needs to be larger than unity in order that a spontaneous current arises. On the other hand, $\gamma$ is proportional to the critical current $I_c$, and this dependence was studied in detail for a d-wave superconductor by many authors [16,17].

In particular Bruder et al [17] calculated the supercurrent tunnel matrix elements in second-order perturbation theory for two d-wave superconductors (1 and 2) with superconducting amplitude

$$\Delta_{d,1/2}(i, T, \phi) = \Delta(i, T) \cos \left[ 2(\phi - \phi_{1/2}) \right],$$

(4)

where $\phi$ is the azimuthal and $\phi_{1/2}$ is the mismatch angle which is here zero because the electronic domains are always aligned with the crystal axis. The dominant contribution to the critical current is from “node to node” tunneling and the overall behavior is like an s-wave superconductor [14]. This result is in agreement with the calculations of a Josephson junction with two different s-wave superconductors [15], which yields a supercurrent proportional to $\Delta_1 \Delta_2/(\Delta_1 + \Delta_2)$.

Bogoliubov-deGennes calculations demonstrate that the values of the local superconducting amplitude also vary locally inside a system with local electronic disorder [19,21]. In Fig.1 we show a typical result of these calculations and plot the temperature evolution of the superconducting amplitude $\Delta_d(i)$ at five randomly chosen different locations or domains $i$.

The disorder of the local gaps implies different values of the tunneling matrix between different grains. Consequently the values of $\gamma$ also change at different junctions ($\gamma(i)$). The values of $\gamma(i)$ increases jointly with $\Delta(i, T)$ as the temperature decreases. Therefore the onset of spontaneous current occurs when the largest $\Delta(i, T)$ reaches a critical value causing some junctions of a certain loop(s) to have $\gamma_{max} \geq 1$ at zero external field, as shown in Fig.3.
Fig. 5: The temperature evolution of the maximum amplitude of the d-wave gap $\Delta(p,T)$ of six different values of average doping $p$. The line at $\approx 40$meV shows the experimental onset of spontaneous magnetization and the intersections yield the temperature onset for different compounds.

Results. – It is very difficult to calculate theoretically the actual values of $\gamma(i)$ but we can obtain some estimation from the experimental data. For instance, it is reasonable that $\Delta(i,T)$ is the major parameter that controls $\gamma(i)$ as the hole doping $p$ is varying. The potential barrier between the grains is important but it is also connected with the values of the superconducting amplitude. Underdoped systems are in general more disordered than the overdoped ones, then it is possible that the number of loops is larger in this limit increasing the strength of the ferromagnetic signal. Indeed the PKE signal observed in underdoped samples are much stronger than those measured near optimally doped [22]. Furthermore, below $T_c$ the phase coherence diminish the phase oscillation causing the signals measured below $T_c$ to be so small that can be almost attributed to oscillation of the data [6,7].

Since the magnetic signal vanishes near $p = 0.18$, we take the onset of $\Delta(i,T)$ that triggers the spontaneous current as the value shown in Fig. 5 of $\Delta_{\text{max}}(p = 0.18, T = 0) \approx 40$meV. In other words, a spontaneous current can exist only above $\Delta \approx 40$meV.

With this maximum value taken as the onset to have $\gamma$ larger than 1, we can follow the temperature evolution of the maximum gap of each compound with the line drawn at $\Delta_{\text{max}}(p = 0.18, T = 0)$ (Fig. 5). The results for 5 doping values are shown in Fig. 5, together with the theoretical results of $\mu$-SR [6] (open circles) and of polar Kerr-effect [7] (black squares).

Conclusions. – In summary, we used the fact that cuprate superconductors have an intrinsic electronic disordered state where the charges are segregated in a few nanometers high and low density grains separated by thin potential walls. As in Bi-clusters [11] this charge confinement may be the origin of the local superconducting interaction. We then calculate the superconducting properties by a BdG method using a phenomenological two-body potential proportional to the energy barriers or walls between the grains [20]. The various distinct regions are coupled forming an array of Josephson junctions that promote the resistivity transition at low temperatures. The phase-number quantum fluctuations are large enough and strong correlation effects promote frustration or negative Josephson coupling [13] that leads to spontaneous currents and an overall magnetization.

This approach connecting an electronic disordered state forming an array of Josephson junctions at low temperatures with frustration effects provides an interpretation to the intriguing spontaneous ferromagnetic phenomenon on cuprates, a long standing problem. This novel calculation shows the importance of the charge inhomogeneities and leads to a pseudogap phase with isolated regions of non-vanishing superconducting amplitudes without long range order.

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