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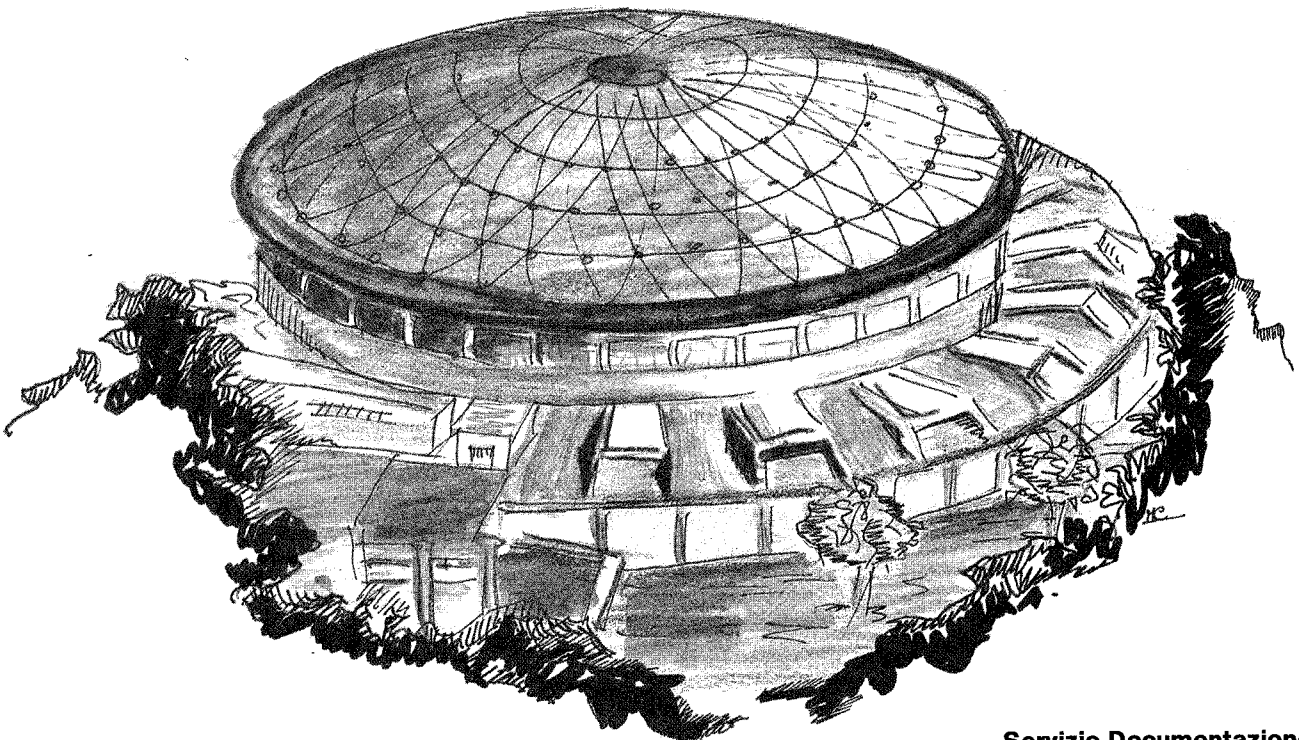
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DIAMAGNETIC PROPERTIES OF SUPERCONDUCTING GRANULAR SYSTEMS

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ABSTRACT

We show that the low field diamagnetic properties of weakly coupled granular high T_c superconductors are dominated by the shielding currents of the whole samples. We remark that, after zero field cooling, by applying a magnetic field to a sample in which a hole is present, a quasi stable non equilibrium condition is reached, so that the critical field $H < \Phi_0 / 2S$ (where S is the hole surface), reported by different authors, loses its meaning. In this non equilibrium condition, the probability of reaching the thermodynamic minimum is determined by the barriers for the input or the output of flux quanta into the superconducting loops which are present in granular samples and are closed by Josephson junctions. Because of fluctuations, the barrier will be exceeded and the junctions will open when the shielding currents reach values near the maximum of the Josephson currents. The apparent lower critical field is determined by the temporary opening of a reasonable high number of junctions which leads to the pinning of flux quanta into the loops. In this way we show that any realistic theoretical model of the Josephson junctions array have to take explicitly into account the shielding current crossing through the junctions.

1. INTRODUCTION

It is well known that the weakly coupled granular structure of high T_C superconductors leads to a fast worsening of both diamagnetic and superconducting transport properties in presence of small magnetic fields. In particular, the resistive transition is a function of the applied measuring current: at higher current values, as the temperature decreases, after a first fall down of the resistance due to the grains transition, a residual resistance generated by the junctions appears. The resistance goes to zero only when the applied current becomes lower than the total value of the magnetic field dependent Josephson currents of the junctions¹. In this case the effects of the current crossing the junctions and of the total field generated by the external sources and by the external and shielding currents are evident. In the same way, in the temperature dependence of the a.c. magnetic susceptibility, a first increase of the diamagnetic signal generated by the superconducting transition of the grains appears as the temperature drops below T_C . A further decrease of the temperature T leads to the superconducting connection between grains and to the complete shielding of the whole system². In an equivalent description, a complete shielding can be measured in the low-field magnetization curve for a constant T lower than T_C . As the magnetic field increases, a lower critical field H_{GC1} dependent on the coupling among the grains and an upper critical field H_{GC2} , above which the grains seem to be decoupled, appears. Finally, since the granular samples are multiply connected topological entities, significant differences in the diamagnetic properties in zero field cooling (ZFC) or field cooling (FC) conditions are detected up to a magnetic field dependent threshold temperature $T^*(H)$ lower than the critical temperature³. In the literature this temperature is identified with the reappearance of a reversible regime in the static ZFC susceptibility or, in a way declared equivalent, to the complete superconducting decoupling of the grains. Recently, it has been proposed that this equivalence may be false⁴. In any case the real nature of both H_{GC1} and H_{GC2} it is not clear in the literature.

From a theoretical point of view some authors explain the magnetic behaviour of such systems in terms of usual flux pinning-flux creep model⁵. On the other hand, many different authors describe the system by a Josephson junctions array model using a spin-glass like Hamiltonian^{6,7}.

In this paper we start from a thermodynamic analysis of superconducting samples in which holes are present. We also remark that the complete shielding of the external magnetic field H leads to quasi stable metastable states. Of course, the system behaviour is dominated by the probability of reaching the thermodynamic minimum. This probability is determined by a potential barrier which has to be exceeded in order to reach the thermodynamic equilibrium. In our case the barriers for the input or the output of flux quanta into the sample is given by the

superconducting loops closed by Josephson junctions which are present in granular samples. For this reason the main well understood results on single loop behaviour are recalled. Starting with this simple approach, we discuss the conditions which lead to a first hysteretic behaviour in the low-field magnetization curve. Following this model, we show that a spin glass like hamiltonian cannot correctly describe granular systems. Conclusions are drawn in the last section.

2. CRITICAL FIELDS OF WEAKLY CONNECTED GRANULAR SYSTEMS

In a granular system a large number of superconducting loops surrounding non superconducting regions and closed by Josephson junctions exists. For this reason some well known features of superconducting rings must be reviewed.

The existence of a lower thermodynamic critical fields H_{GC1} means that the absolute minimum of the correct thermodynamic potential $G(T,H)$, for $H < H_{GC1}$, should correspond to the complete shielding state. In the presence of an external magnetic field H , it is well known that the density $\mathcal{G}(T,H)$ of the thermodynamic potential is given by the equation ⁸:

$$\mathcal{G}(T,H) = \mathcal{G}_0(\Delta(x,y,z,T)) + (h - H)^2/8\pi \quad 1$$

where $\Delta(x,y,z,T)$ is the order parameter and h is the local value of the magnetic field. By eq.1 it is evident that, in the presence of non superconducting regions inside the ring, the absolute minimum of $G(T,H)$ can be found for $h = H$ inside the non superconducting regions⁹, so that the value of the lower critical field is zero. The flux quantization condition $\Phi(B) = n\Phi_0 = BS$ (where S is the hole surface, Φ_0 is the flux quantum, n is an integer and B is the mean value of h) gives restrictions, so that only states with values of $B = n\Phi_0/S$ inside the hole exist. In this way, if the magnetic field is roughly constant in the hole, the thermodynamic potential can be approximated by:

$$G(T,H) = G_R(T,H) + \gamma(n\Phi_0 - SH)^2 \quad 2$$

where $G_R(T,H)$ is the thermodynamic potential of the superconducting part of the ring and γ is a geometrical factor. In the "pure superconducting state" $d\Phi(B)/dt=0$, so that n is constant despite the fact that H may vary. By eq.2 it is clear that, starting by ZFC condition ($n=0$) and applying a magnetic field H at $T < T_c$ to a multiply connected superconductor, the complete shielding state ($n=0$) will correspond to the absolute minimum of $\mathcal{G}(T,H)$ only if the following condition is met:

$$\mathbf{H} < \mathbf{H}_{hC1} = \Phi_0 / 2 S$$

On the contrary for $\mathbf{H} > \mathbf{H}_{hC1}$ the minimum of $G(T, \mathbf{H})$ corresponds to the presence of flux quanta in the hole ⁶. In a second type superconducting ring this state is reached only by means of the intermediate state in which a flux quantum is present in the superconducting region of the ring. Below \mathbf{H}_{C1} of the bulk material, the increase of $G(T, \mathbf{H})$, corresponding to this intermediate state, is so high to make completely negligible the probability of reaching the thermodynamic minimum. In this way the non equilibrium state is stable, so that flux quanta do not enter inside the hole also for $\mathbf{H} \gg \mathbf{H}_{hC1}$. Therefore, \mathbf{H}_{hC1} loses its meaning. Similar arguments can be used in case of superconducting rings closed by Josephson junctions, but in these cases fluxoid quantization must be taken into account. These configurations have been extensively studied for SQUID applications with an excellent agreement between experimental results and theoretical predictions ⁸. In this case the flux quanta prefer to enter in the hole through the junction generating a 2π flip of the junction phase ϕ . This phenomenon determines the energy barrier which must be overcome to reach the stable state. For an isolated small Josephson junction, in the absence of bias current, the hamiltonian is⁸: $\mathcal{H} = I_j(\mathbf{h}) (h/2e) (1 - \cos\phi)$, (where $I_j(\mathbf{h})$ is the maximum value of the superconducting current in the local magnetic field), so that the barrier height for the flux quanta is $\Delta_E = I_j(\mathbf{h}) (h/e)$.

However it is well known that, in the presence of a bias current I_B , the correct hamiltonian is: $\mathcal{H} = I_j(\mathbf{h}) (h/2e) (1 - \cos\phi + \alpha\phi)$, where $\alpha = I_B / I_j(\mathbf{h})$. The presence of the term $\alpha\phi$ introduces a slope on the hamiltonian which, removing the state degenerations, strongly changes the system dynamic. The barrier height is reduced and it is given by:

$$\Delta_E = I_j(\mathbf{h}) (h/e) \{ (1-\alpha^2)^{1/2} - \alpha \cos^{-1}\alpha \} \quad 4$$

In this case Δ_E goes to zero as I_B reaches $I_j(\mathbf{h})$.

The same result is valid for junctions closing rings: in this case the bias current is the shielding current I_S , which decreases as flux quanta enter in the ring. In this way, for a fixed external magnetic field the thermodynamic potential is given by the equation ⁸:

$$G = (\Phi - \Phi_{EX})^2 / 2L + I_j(\mathbf{h}) (h/2e) \{ 1 - \cos(2\pi\Phi/\Phi_0) \} \quad 5$$

Where L is the loop inductance, and Φ_{EX} is the flux of the external magnetic field. Equation 5 is the superimposition of the oscillating cosine term to the negative slope depending on the shielding current. Due to the presence of thermal fluctuations, the superconducting ring will open (from the superconducting point

of view) and flux quanta enter into the ring for values of shielding currents just near the maximum of the Josephson current, as the condition $\Delta_E \approx kT$ is verified. The number of flux quanta, which actually goes inside the ring, is function of the ring and junction parameters⁸. In any case during the temporary opening of the ring, the circulating current decreases, until the ring closes again.

In granular systems the presence of a network, made of a large number of superconducting loops closed by Josephson junctions, generates a complex behaviour. However the main results obtained for a single loop are still valid. After ZFC, the application of a magnetic field generates shielding currents circulating in the outer sheath of the sample through a large number of junctions. The shielding currents will open the weakest junction of the outer sheath, when the maximum value of Josephson current of the junction is reached. In this way flux quanta enter in an internal loop just underneath the sample surface. A further increase of the magnetic field leads at the beginning to the opening of stronger junctions of the outer sheath and later, to the opening of internal shells. In this way a progressive magnetic field penetration is obtained. More details on the dynamics of flux quanta penetration is reported by A. Saggese et al.¹⁰. In any case the apparent lower critical field of the granular system H_{GC1} is not well defined and it is determined by the opening of a reasonable high number of junctions, which leads to the penetration and to the pinning of flux quanta into the loops.

In any case the hamiltonian of the whole system must be written as:

$$\mathcal{H} = \sum_n I_{j_n}(\mathbf{h}) (h/2e) (1 - \cos\varphi_n - \alpha_n \varphi_n) \quad 6$$

where φ_n is the gauge invariant phase of the n-th junction and α_n is the ratio between the current crossing through the n-th junction and the maximum value of the superconducting Josephson current which can go through the junction in the presence of the local magnetic field \mathbf{h} . In this way we believe that the dynamics of the system described by eq.6, due to the presence of the linear term, is qualitatively different from a spin glass system. Indeed, as previously noted, the linear term removes the degeneration of the states and reduces to zero the barrier heights. Moreover, since eq.1 is still valid, any 2π variation of φ corresponds to the input or output of a flux quantum into a loop, in such way that \mathbf{h} goes toward \mathbf{H} and $G(\mathbf{T}, \mathbf{H})$ is minimized.

Following this model, as described by S.Pace et al.¹¹, a correct description of the Josephson junction array, generated by the granular system, leads to a model very similar to the usual flux pinning - flux creep model. In our case the pinning centers are the non superconducting regions closed by the loops and the pinning potential is given by the Josephson $\cos \varphi$ term. In the usual systems the presence

of currents generates Lorentz forces, proportional to the current, which reduces the pinning potential. In an analogous way in our description the potential barriers are reduced directly by the currents crossing the junctions.

3. CONCLUSIONS

Extending the well established results obtained for superconducting rings containing one junction to a network of junctions, our model predicts:

- a) the absence of a lower thermodynamic critical field H_{GC1} ,
- b) the experimental presence of a lower critical field, determined by the I_{S_n} current crossing the n-th junction, and the maximum Josephson current $I_{j_n}(h)$ in the local magnetic field h .

Following this description any model constructed to describe the diamagnetic behaviour of a superconducting granular system must take explicitly into account the presence of shielding currents. On the contrary superconducting glass models neglect such currents. We believe that such approximation changes qualitatively the system dynamic. On the contrary a correct description of a granular system as an ensemble of superconducting loops leads to a description similar to the traditional flux creep model.

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