Spin-size disorder model for granular superconductors with charging effects

Enzo Granato [a,*] and Giancarlo Jug [b]

[a] *Laboratório Associado e Sensores e Materiais, Instituto Nacional de Pesquisas Espaciais, 12227-010 São José dos Campos, SP Brazil*
[b] *Dipartimento di Fisica e Matematica, Università dell'Insubria, Via Valleggio 11, 22100 Como, Italy*
CNISM – Unità di Ricerca di Como and INFN – Sezione di Pavia, Italy

Abstract

A quantum pseudo-spin model with random spin sizes is introduced to study the effects of charging-energy disorder on the superconducting transition in granular superconducting materials. Charging-energy effects result from the small electrical capacitance of the grains when the Coulomb charging energy is comparable to the Josephson coupling energy. In the pseudo-spin model, randomness in the spin size is argued to arise from the inhomogeneous grain-size distribution. For a particular bimodal spin-size distribution, the model describes percolating granular superconductors. A mean-field theory is developed to obtain the phase diagram as a function of temperature, average charging energy and disorder.

Key words: Granular superconductors, Josephson-junction arrays, Superconductor-insulator transition
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1. Introduction

Quenched disorder is obiquitous in condensed-matter systems, sometimes determining entirely new physical properties and phenomena. For the theoretical explanation of many experimental findings, models of classical and quantum spin systems are employed and - typically - disorder is introduced through a spatial variation of the exchange couplings, of the direction of the axial anisotropies, and of the sign of the couplings. Theoretical and experimental studies of model and real physical systems with these kinds of disorder have lead to much new physics. Much discussed have been the glassy phases in magnetic [1,2,3], and superconducting materials [4].

In superconductors, specially with the advent of high-T_c ceramic superconductors, the role of disorder has become central in the discussion of the physical properties of real materials [3,4]. Many materials are naturally microstructured and granularity characterises the mesoscopic structures of most systems, leading to a phase diagram often displaying a superconductor-insulator transition at zero temperature due to the charging energy of the grains. As first pointed out by Abeles [5], when the grain charging energy arising from the charge Q and capacitance C of the grain, $E_c = Q^2/2C \sim (2e)^2/d$ (d is the grain diameter and e the electronic charge) is larger than the Josephson-coupling energy E_o between nearest neighbors grains, phase coherence is destroyed due to zero-point quantum fluctuations of the local phase of the superconducting order parameter [6]. Granularity can also be realized in a controlled manner in artificially fabricated Josephson-junction arrays of coupled superconducting grains [7], with a space dimensionality d less than 3. Such granular systems can be theoretically modelled by pseudo-spin systems, the 'spins' representing with their states the few relevant quantum charge states of the grains at relatively low temperatures. In such models the pseudo-spin size is the same for all grains corresponding to the assumption that the grain-size distribution is very narrow, or alternatively, of negligible charging-energy disorder. A well known model of this type is the pseudo-spin-one model introduced by de Gennes and studied in different works [3,8,9,10] where only charge states $-1, 0, 1$ are allowed, corresponding to $S = 1$. However, realistic systems may contain different kinds of disorder, such as a spatial distribution of Josephson couplings between grains or/and a distribution of grain sizes, which leads to disorder in the grain electrical capacitances and charging energies.

Studies of the effects of disorder in the electrical capacitance or charging energy of the grains have appeared recently [11,12,13]. Within a mean-field approximation [11], charging energy disorder widens the extent of the super-

* Corresponding author. Tel/Fax: +55 12 39456717
Email address: enzo@las.inpe.br (Enzo Granato).

conducting phase at the expense of the insulating one. This behavior is consistent with results for the superconductor-insulator transition from quantum Monte Carlo simulations in two-dimensional models of Josephson-junction arrays [13] with disorder in the diagonal capacitance matrix. Earlier calculations for the related boson Hubbard model with disorder in the onsite Coulomb repulsion [14] are also consistent with a decrease in the phase coherence threshold and moreover suggest a different universality class from the non-disordered case [15].

In this work, we consider the effects of charging energy disorder in granular superconducting materials within the pseudo-spin approach. By considering a spatial distribution of the grain sizes, which leads to local charging energy disorder, a quantum pseudo-spin model with random on-site spin sizes can be constructed. A mean-field theory is developed to obtain the phase diagram as a function of temperature, average charging energy and disorder.

Spin-size disorder models have seldom been considered in the literature. In the context of classical spin models, spin-size disorder can be readily turned into exchange-coupling disorder; the physics of quantum systems with spin-size disorder, however, appears to have not been investigated in such depths. It has been considered mostly within the one-dimensional systems: dilution of a quantum spin-$\frac{1}{2}$ 2-ladder was studied by Sigrist and Furusaki [1] whilst the general problem of a quantum spin chain with random S as well as random J was considered by Westerberg *et. al* [2] within a real-space renormalization-group method showing that these systems belong to a different universality class of disordered spin systems.

2. Pseudo-spin model with random spin sizes

The Hamiltonian for a set of superconducting grains coupled by the Josephson energy can be written as the sum of the Coulomb charging energy and the Josephson-coupling energy [3,6,8]

$$H_{gs} = \frac{1}{2}\sum_{i,j} C_{ij}^{-1} Q_i Q_j - \sum_{<ij>} E_{ij}\cos(\theta_i - \theta_j), \qquad (1)$$

where Q_i is the net charge on the superconducting grain at site i and θ_i is the phase of the local superconducting order parameter ψ_i. C_{ij} is the electrical capacitance matrix and E_{ij} is the Josephson coupling between nearest-neighbor grains. The charge Q_i in each grain can be expressed in terms of the excess number of Cooper pairs n_i as $Q_i = 2en_i$. Considering a diagonal capacitance matrix $C_{ij} = C_i\delta_{ij}$ and uniform Josephson-coupling energies $E_{ij} = E_o$, leads to the self-charging model

$$H_{gs} = 2\sum_i U_i n_i^2 - E_o \sum_{<ij>} \cos(\theta_i - \theta_j), \qquad (2)$$

where $U_i = e^2/C_i$ are the charging energies of the grains. The number operator n_i is conjugate to the phase θ_i, satisfying the commutation relation

$$[n_i, \theta_j] = -i\delta_{ij}, \qquad (3)$$

and can be written as $n_i = -i\partial/\partial\theta_i$, having integer eigenvalues $0, \pm1, \pm2....$ The model of Eq. (2) can also be regarded as a boson Hubbard model [14,15] where the charging energy represents the onsite Coulomb repulsion of bosons and the Josephson coupling represents the hopping term.

When the charging energies are uniform $U_i = U_o$, a pseudo-spin one model for the Hamiltonian of Eq. (2) can be constructed by truncating the basis vectors of the number operators $|n_i>$ to $|0>$ and $|\pm 1>$, corresponding to the lowest charging energy states. Identifying the charging states, $|n_i>$, as the eigenstates of S^z for spin $S = 1$, writting the second term of Eq. (2) in terms of $e^{\pm i\theta_i}$ and making the correspondence

$$e^{i\theta_i} \to S_i^+/\sqrt{2}, \qquad e^{-i\theta_j} \to S_j^-/\sqrt{2}, \qquad n_i \to S_i^z, \qquad (4)$$

one obtains the de Gennes mapping to a $S = 1$ pseudo-spin model with single-ion spin-anisotropy [3,8]

$$H_{S=1} = 2U_o\sum_i (S_i^z)^2 - \frac{E_o}{4}\sum_{\langle ij\rangle}(S_i^+ S_j^- + S_j^+ S_i^-), \qquad (5)$$

with $S_i^+ = S_i^x + iS_i^y$. The factor $1/\sqrt{2}$ in front of $S^{\pm}$ in Eq. (4) takes into account the length of the pseudo spin $\sqrt{S(S+1)}$. In terms of this spin model, the superconductor to insulator transition in the original granular superconductor corresponds to a zero temperature transition where the ferromagnetic order of the spins in the xy plane is destroyed by quantum fluctuations when the S^z component is confined to zero for an increasing ratio of U_o/E_o. Although this mapping is not exact [18], the $T = 0$ critical behavior observed in the original phase model of Eq. (2) without disorder and the spin model is the same [14,19] as found in numerical calculations in one dimension. For higher dimensions, results from the same mean-field approximation applied to both models also agree [8].

To consider the main effects of disorder in the charging energies U_i, we generalize the above approximate mapping to a pseudo-spin model with randomness in the spin-size values S_i. Since for a given charging energy (or temperature fluctuation), low values of U_i correspond to charging states with higher n_i, it seems reasonable to use a truncation scheme which identifies the charging states $|n_i>$ and $|n_j>$ with the S^z-eigenstates of the spins of different sizes S_i and S_j, such that the corresponding maximum charging energies $2U_i\mathcal{S}_i^2$ and $2U_j\mathcal{S}_j^2$ of the grains are both comparable to the same truncation energy (here $\mathcal{S}_i = \sqrt{S_i(S_i+1)}$ is the length of the spin $\mathbf{S}_i$). This (approximate) mapping leads to the effective spin Hamiltonian

$$H = D\sum_i \frac{1}{\mathcal{S}_i^2}(S_i^z)^2 + J\sum_{\langle ij\rangle}\frac{1}{\mathcal{S}_i\mathcal{S}_j}(S_i^+ S_j^- + S_j^+ S_i^-), \qquad (6)$$

with randomness in the spin-size values S_i. Here, $J = -E_o/2$, $D = 2\bar{U}\bar{\mathcal{S}}^2$ where $\bar{U}$ is the average value of U_i and

$\bar{\mathcal{S}}$ the average value of $\mathcal{S}_i$. The spin sizes S_i are restricted to take only integer values $0, 1, 2, 3, ...$ in this mapping. In Eq. (6), randomness in the spin-size also leads to randomness in the local single-ion anisotropy parameter, but these random variables are only correlated at the same site.

A typical spin-size distribution we shall be considering is

$$P(S_i) = x \sum_{L=\pm 1} \delta(S_i - S_o - L) + (1-2x)\delta(S_i - S_o), \quad (7)$$

where three spin-size values S_o, S_o+1 and S_o-1 are mixed with average value S_o and a concentration x, which is a measure of the disorder. It is also convenient to rewrite the spin model of Eq. (6) in terms of normalized spins $\tilde{S}_i = S_i/\mathcal{S}_i$

$$H = D\sum_i (\tilde{S}_i^z)^2 + J\sum_{\langle ij \rangle} (\tilde{S}_i^+ \tilde{S}_j^- + \tilde{S}_j^+ \tilde{S}_i^-). \quad (8)$$

In this latter form, a bimodal distribution of spin values can be considered:

$$P(\tilde{S}_i) = x\delta(\tilde{S}_i - \tilde{S}_1) + (1-x)\delta(\tilde{S}_i - \tilde{S}_2), \quad (9)$$

which in the special case $\tilde{S}_1 = 0$ corresponds to the dilution of a spin-S_2 system or to percolating granular superconductors [16] in the presence of charging effects.

3. Mean-Field Theory

For high enough space dimensions d, reasonable results can be obtained from simple mean-field approximations (MFA). The MFA replaces all variables around a given small cluster with their average value, or order parameter, and then use the resulting approximate Hamiltonian to evaluate the order parameter itself. Since the spin sizes are random, one must carry out the averaging procedure with respect to $P(S_i)$ as well and we denote this by $[...]_{ave}$.

The simplest cluster is a single spin on a site. With $\mathbf{M} = [< \mathbf{S}_i >]_{ave}$ the mean field, this yields a self-consistent equation for the order parameter $\mathbf{M}$:

$$\mathbf{M} = [\langle \mathbf{S}_i \rangle]_{ave} = \left[\frac{1}{Z_{MFA}} Tr_{\mathbf{S}_i} \mathbf{S}_i e^{-\beta \bar{H}_{MFA}} \right]_{ave}, \quad (10)$$

where $\bar{H}_{MFA}$ is the mean-field Hamiltonian and $\beta = 1/k_B T$. For the anisotropic Hamiltonian of Eq. (6), we assume that the ferromagnetic ordering takes place in the xy plane of the anisotropy, say $[< S_i^x >]_{ave} = M$. Then, in the MFA we replace [3]

$$S_i^x S_j^x + S_i^y S_j^y \to \langle S_i^x \rangle S_j^x + S_i^x \langle S_j^x \rangle - \langle S_i^x \rangle \langle S_j^x \rangle, \quad (11)$$

by virtue of the fact that $< S_i^y >= 0$. Near the critical curve, the MFA Hamiltonian then becomes:

$$\bar{H}_{MFA} = D\sum_i ((\tilde{S}_i^z)^2 - \lambda \tilde{S}_i^x) + O(\tilde{M}^2), \quad (12)$$

where $\lambda = 2z|J|\tilde{M}/D$ is a dimensionless expansion parameter.

Since we are interested in the phase transition line only, we can assume $\tilde{M} \equiv M/\mathcal{S}$ small and use a first-order perturbation expansion to solve Eq. (10) for arbitrary spin S. In a now single-site problem, we reabsorb a factor $\mathcal{S}^{-2}$ into D and J. The unperturbed states $|m>$ are eigenstates of S^z and thus also of the unperturbed Hamiltonian $H_o = D(S^z)^2$, with $m = -S, -S+1, ..., S-1, S$. We have

$$\langle S^x \rangle = \frac{\sum_m e^{-\beta E_m} \langle \psi_m | S^x | \psi_m \rangle}{\sum_m e^{-\beta E_m}}, \quad (13)$$

where to first-order $E_m = Dm^2 + \langle m| - \lambda D S^x |m\rangle = Dm^2$ and

$$|\psi_m\rangle = |m\rangle + {\sum_{m',m}}' \frac{\langle m'| - \lambda D S^x |m\rangle}{E_m^0 - E_{m'}^0} |m'\rangle$$
$$= |m\rangle - \lambda {\sum_{m',m}}' \frac{\langle m'|S^x|m\rangle}{m^2 - m'^2} |m'\rangle. \quad (14)$$

Here, the prime on $\sum'$ means $m' \neq m$. These can then be inserted in Eq. (13) to get, keeping only first order terms in λ:

$$\langle S^x \rangle = \frac{2}{Z_0} {\sum_{m',m}}' e^{-\beta D m^2} \frac{\langle m|S^x|m'\rangle\langle m'|S^x|m\rangle}{m'^2 - m^2} \lambda, \quad (15)$$

where $Z_0 = \sum_{m=-S}^{S} e^{-\beta D m^2}$. Using

$$\langle m|S^x|m'\rangle = \frac{1}{2}(A_{m-1}^S \delta_{m',m-1} + A_m^S \delta_{m',m+1})$$
$$A_m^S = \sqrt{(S-m)(S+m+1)}, \quad (16)$$

we get

$$\langle S^x \rangle = \frac{\lambda}{2Z_0} \sum_m \{ \frac{(A_m^S)^2}{(m+1)^2 - m^2} + \frac{(A_{m-1}^S)^2}{(m-1)^2 - m^2} \} e^{-\beta D m^2} \quad (17)$$

which, using Eq. (10), gives implicitly the critical temperature in the MFA

$$\frac{D}{2z|J|} = \Big[\frac{1}{\sum_m e^{-Dm^2/(S(S+1)k_B T_c)}} \sum_{m=-S}^{S} \frac{S(S+1)+m^2}{1-4m^2} e^{-Dm^2/(S(S+1)k_B T_c)} \Big]_{ave} \quad (18)$$

One can check that for $S = 1$ and in absence of disorder the MFA transition line from Eq. (18) agrees with the result obtained for the de Gennes model of Eq. (5) in the same approximation [3]. Fig. 1 shows the phase boundaries between the superconducting and normal phases obtained in absence of disorder for different values of uniform spin sizes $S = 1$, $S = 2$ and $S = 5$. The critical value E_o/U_o for the superconductor-insulator transition at $T = 0$ remains unchanged and only at much higher temperatures there is a deviation in the transition lines when considering larger spin approximations. This is in agreement with MF calculations using truncated basis vectors in the phase model of Eq. (2) for increasing number of states [8].

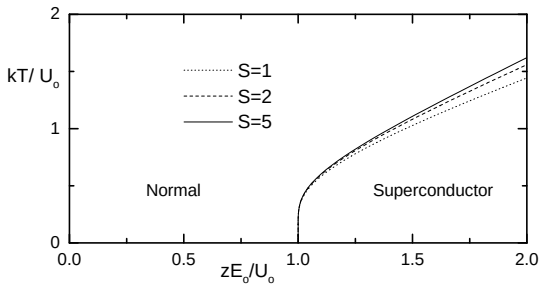

Fig. 1. Phase boundary without disorder obtained from the pseudo-spin model with different uniform spin values $S_i = 1, 2$ and $S_i = 5$.

Our present calculations generalize the MFA to arbitrary spin sizes and allow to obtain the transition line in the case of a random spin size distribution by simply averaging with respect to $P(S)$ in Eq. (18). The consequences of spin-size disorder depend very much now on the chosen distribution, but it is expected that in general within a simple MF approximation, the phase transition of the homogeneous system is preserved with at most a modification of the phase boundary line. Fig. 2a shows the transition lines for the spin-size distribution of Eq. (7) with $S_o = 2$ and different values of the disorder parameter x. For increasing disorder the critical value for phase coherence $E_o/\bar{U}$ decreases, increasing the extent of the superconducting phase at low enough temperatures. This behavior is in qualitative agreement with MF calculations [11] in the phase model of Eq. (2).

On the other hand, a different behavior is expected when there is dilution of superconducting grains. Fig. 2b shows the transition lines for the spin-size distribution of Eq. (9) with $\tilde{S}_1 = 0$, $S_2 = 2$, corresponding to a dilution of grains for different concentrations x. In this case, disorder decreases the extent of the superconducting phase. For large values of x, corresponding to the system below the percolation threshold, the superconducing phase should disappear. However, an improved MF approximation is required to describe this behavior. One possible approach is the MF renormalizaton-group method [17] used previously to study the de Gennes model of Eq. (5) in absence of disorder [9].

4. Conclusions

We have introduced a quantum pseudo-spin model with random spin sizes to model the effects of charging-energy disorder in granular superconducting materials. Randomness in the spin size is argued to arise from the inhomogeneous grain-size distribution. For a particular bimodal spin-size distribution, the model describes percolating granular superconductors. A mean-field theory has been developed to obtain the phase diagram as a function of temperature, average charging energy and disorder. The results are qualitatively consistent with previous mean-field calculations in the phase-number representation. The pseudo-spin model should provide a useful framework to study the critical behavior and universality classes in presence of strong charging-energy disorder.

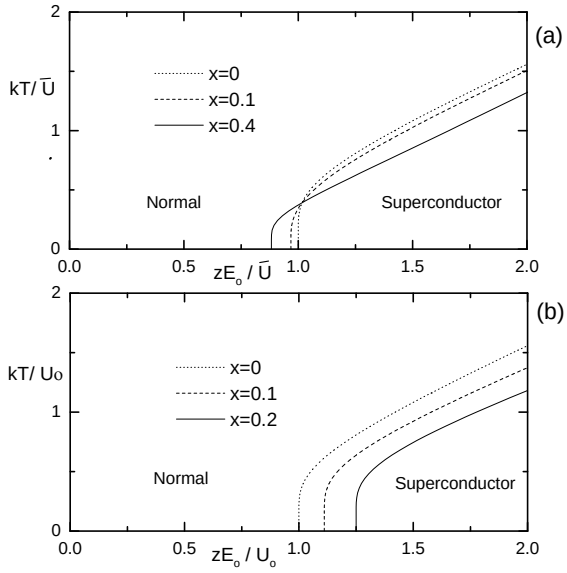

Fig. 2. Phase boundary with disorder obtained from the pseudo-spin model with random spin sizes: a) with charging energy disorder, corresponding to disordered spins values giving by the probability distribution of Eq. (7), with average spin value $S_o = 2$ and different x ; b) with dilution of grains, corresponding to the disordered spin values with the probability distribution of Eq. (9), with $\tilde{S}_1 = 0$ and $S_2 = 2$ and different x

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References

[1] M. Sigrist and A. Furusaki, J. Phys. Soc. Jpn **65**, 2385 (1996).
[2] E. Westerberg, A. Furusaki, M. Sigrist and P.A. Lee, Phys. Rev. Lett. **75**, 4302 (1995); *ibid.* Phys. Rev. B 55, 12578 (1997).
[3] E. Simánek, *Inhomogeneous Superconductors: Granular and Quantum Effects* (Oxford UP, New York 1994)
[4] D.S. Fisher, M.P.A. Fisher, and D. Huse, Phys. Rev. B **43**, 130 (1991).
[5] B. Abeles, Phys. Rev. B **15**, 2828 (1977).
[6] S. Doniach, Phys. Rev. B **24**, 5063 (1981).
[7] R. Fazio and H. van der Zant, Phys. Rep. **555**, 235 (2001).
[8] P. Fazekas, B. Muhlschlegel, and M. Schroter, Z. Phys. B **57**, 193 (1984).
[9] E. Granato and M.A. Continentino, Phys. Rev. B **48**, 15977 (1993).
[10] T.K. Kopeć, Phys. Rev. B **69**, 054504 (2004).
[11] F.P. Mancini, P. Sodano and A. Trombettoni, Phys. Rev. B **67**, 014518 (2003).
[12] W.A. Al-Saidi and D. Stroud, Phys. Rev. B **67**, 024511 (2003).
[13] W.A. Al-Sadi and D. Stroud, Physica C **402**, 216 (2004).
[14] M.-C. Cha and S.M. Girvin, Phys. Rev. B **49**, 9794 (1994).
[15] M.-C. Cha, M.P.A. Fisher, S.M. Girvin, M. Wallin, and A.P. Young, Phys. Rev. B **44**, 6883 (1991).
[16] S. John and T.C. Lubensky, Phys. Rev. Lett. **55**, 1014 (1985).
[17] J.O. Indekeu, A. Maritan, and A. Stella, J. Phys. A **15**, L291 (1982).
[18] D. Das and S. Doniach, Phys. Rev. B **60**, 1261 (1999).

[19] U. Glaus, Physica **141**A, 295 (1987).