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Coupled two-order-parameter approach to granular superconductors

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A two-order-parameter approach to the theory of granular superconductors is presented, in which the paracoherent transition and the superconductive one are treated accounting for their mutual coupling. The appearance of a tricritical line in the phase diagram is discussed. Renormalization of the amplitude of the order parameter is predicted in the whole temperature range, leading to a satisfactory understanding of experimental data previously unexplained.

It is by now a well-established fact that strongly inhomogeneous superconductors (granular superconductors) can be modeled by arrays of coupled Josephson junctions.[1] The currently employed model Hamiltonian contains a charging term[2] besides the usual intergrain Josephson coupling term E_J; more recently dissipative effects due to the coupling with a thermal bath[3] and/or to quasiparticle tunneling[4] have been introduced into the theory. From the most recent studies, at least three main features appear to be emerging: first, the possibility of a reentrant behavior of the critical temperature T_J,[5] the temperature at which the system undergoes the paracoherent transition; second, the feasibility of a Kosterlitz-Thouless-Berezinskii transition in systems of restricted geometries,[6] and third, the role of dissipation.[7]

In the usual approach to the study of the paracoherent transition, the phase ϕ of the superconducting order parameter of the grains Δ is the only relevant dynamical variable. This picture is suitable, for example, when T_J is much lower than the single-grain superconducting transition temperature T_C. In the granular Al samples studied by Shapira and Deutscher,[8] however, this two-step behavior is not observed showing that $T_J \ll T_C$ cannot be taken as a rule.

In such systems it seems more appropriate to treat the two transitions on the same footing; this implies that we cannot any longer disregard the $|\Delta|$ dependence of E_J. This leads to a coupling between the two order parameters $|\Delta|$ and Ψ which describe, respectively, the onset of the single-grain superconductivity and the locking of the phases in the array.

The aim of this article is to present the appropriate free energy for dealing with the case $T_J \lesssim T_C$ and to show how, even in the simplest evaluation scheme, the coupling between $|\Delta|$ and Ψ provides a richer picture. We will show, for example, that the paracoherent transition is likely to be first order for suitable sizes grains.

The treatment of Ambegaokar, Eckern, and Schön (AES) can be generalized for our purposes. We express the partition function as

$$\mathcal{Z} = \int \mathcal{D}\Delta\, \mathcal{D}\phi \exp[-\mathcal{S}(\Delta,\phi)/\hbar] , \tag{1}$$

where

$$\mathcal{S} = -\sum_i \mathrm{Tr}\ln(G_i^{(0)-1}) + \lambda^{-1}\sum_i \int d^3r |\Delta_i(\mathbf{r})|^2 + \int_0^{\beta\hbar}\left[\sum_i (\tfrac{1}{2})U^{-1}\hbar^2 \left[\frac{\partial\phi i}{\partial\tau}\right]^2 + \sum_{ij} E_J\{1-\cos[\varphi_{ij}(\tau)]\}\right] + (\tfrac{1}{2})\int_0^{\beta\hbar} d\tau\, d\tau' \sum_{ij}\alpha(\tau-\tau')\sin^2\{[\phi_{ij}(\tau)-\varphi_{ij}(\tau')]/2\} \tag{2}$$

and $G_i^{(0)}$ is the Nambu-Gor'kov matrix Green's function of the ith grain, λ is the Bardeen-Cooper-Schrieffer (BCS) coupling constant, U is the charging energy $\alpha(\tau-\tau')$ is the kernel describing the dissipation spectrum, and the integral in the second term refers to the grain volume.

The first two terms in Eq. (2) refer to the single grains, while the remaining ones describe, respectively, charging, Josephson, and dissipation effects.

The need of retaining the full functional dependence of $\mathcal{S}$ on $|\Delta|$ requires the inclusion of the first two terms in Eq.

(2), this last not appearing in Eq. (32) of Ref. 7. In fact the AES approach rests on the assumption that the single grain (as, for example, the mean-field value of $|\Delta|$) and the coherence properties of the array are decoupled because the single-grain condensation energy is large on the scale of E_J. Then AES evaluate $|\Delta|$ from the first two terms in (2) while the remaining becomes the well-celebrated effective phase action for the granular system. When the above assumption is not valid (as, for example, when $T_J \lesssim T_C$) a generalization of the AES treatment is in order.

Performing the Hubbard-Stratanovich transformation to the phase-dependent part of $\mathcal{S}$ (Refs. 1 and 9) (which introduces the auxiliary complex field Ψ, see Ref. 1), and expanding the first two terms of Eq. (2) in powers of $|\Delta|$, we present, in the continuum limit, the following expression for the partition function $\mathcal{Z}=\int \mathcal{D}|\Delta|\mathcal{D}\Psi \exp(-\mathcal{F})$, where

$$\mathcal{F}(|\Delta|,\Psi)=V_\rho^{-1}\int d\mathbf{r}\left[a|\Delta(\mathbf{r})|^2+\frac{b}{2}|\Delta(\mathbf{r})|^4+\beta^{-2}\sum_n[1-c_n|\Delta(\mathbf{r})|^2]\Psi_n(\mathbf{r})|^2 + (1/2\beta^4)d|\Delta(\mathbf{r})|^4\sum_{n_i}\Psi^*_{n_1}(\mathbf{r})\Psi_{n_2}(\mathbf{r})\Psi^*_{n_3}(\mathbf{r})\Psi_{n_4}(\mathbf{r})\delta\left[\sum_i n_i,0\right]+\beta^{-2}g|\Delta(\mathbf{r})|^2\sum_n|\nabla_r\Psi_n(\mathbf{r})|^2\right], \quad (3)$$

where the coefficients are

$$a=-\delta^{-1}(1-T/T_C)(\beta_J/\beta_c),$$

$$b=[7\zeta(3)/8\pi^2]\delta^{-1}(\beta_J/\beta_c),$$

$$c_n=(\tfrac{1}{2})z(R_0/R_N)(\beta_J/\beta_c^2)w(\omega_n),$$

$$d=(\tfrac{1}{4})[z(R_0/R_N)(\beta_J/\beta_c^2)w(0)]^2,$$

$$g=(\tfrac{1}{4})z(R_0/R_n)(\beta_J/\beta_0^2)w(0)\rho^2,$$

with

$$w(\omega_n)=\int_0^{\beta\hbar}d\tau\langle e^{i\phi(\tau)}e^{i\phi(0)}\rangle_0\exp(-i\omega_n\tau),$$

$$\Psi(\mathbf{r},\tau)=\beta^{-1}\sum_n\Psi_n(\mathbf{r})\exp(i\omega_n\tau),$$

$$\omega_n=2\pi n/\beta\hbar,\quad n=1,2.$$

Here $\delta=\beta_c[N(0)\Omega]^{-1}$, $N(0)$ being the electron density of states at the Fermi surface and Ω the volume of the grain, $\zeta(3)$ is the Riemann ζ function, z is the lattice coordination number, $R_0=h/4e^2$, R_N is the normal sheet resistance, ρ is the lattice constant, V_ρ is the volume of the elementary cell, $|\Delta|$ is chosen to be dimensionless via the transformation $\beta_c|\Delta|\rightarrow|\Delta|$, and $\langle\,\rangle_0$ refers to the average over charging and dissipative part of the action. The integration in (3) is the continuous limit for the lattice summation, i.e., $\sum\rightarrow\int$.

The previous expression for the free energy was derived under the following assumptions.

(i) Due to the smallness of the grains, $|\Delta|$ is taken uniform inside the grain [the $\mathbf{r}$ dependence in Eq. (3) takes into account only long-wavelength variations from grain to grain].

(ii) Quasiparticle excitations are entirely neglected, whereas the finiteness of the grain size is accounted for via the parameter δ.[10]

(iii) The Josephson coupling is evaluated neglecting $0(|\Delta|^6)$.

(iv) Quantum fluctuations in $|\Delta|$ are neglected.

The obtained free-energy functional can be viewed as a novelty; indeed it is worth noticing that although a similar expression was derived elsewhere[11] by a different method, the functional approach here developed allows to introduce just from the outset both charging and dissipative effects.

The coupling between the two order parameters $|\Delta|$ and $|\Psi|$ is of the kind $|\Delta|^2|\Psi|^2$. This can be understood noticing that for T_J close to T_C we can use the limiting form of the Ambegaokar-Baratoff expression of E_J for $|\Delta|\rightarrow 0$. Moreover in this region the static ansatz for $|\Delta|$ is appropriate. Even if for the high-temperature-limit results presented in this article Ψ can be considered as classical too, we retain its τ dependence because the present expression is feasible to further investigate the effect of charging in the semiclassical region.[12]

The coupling between a complex field (in our case Ψ) and another scalar ordering field (Δ) becoming critical at a different temperature has been extensively treated into the literature[13] for the understanding of the interplay of different modes of ordering. The situation here is somewhat more complicated because we do not deal with the real coexistence of two orderings due to the fact that the paracoherent transition is unthinkable without the previous onset of local superconductivity. This is reflected in the peculiar form of the obtained free energy.

Even in the mean-field approximation for $|\Delta|$ novel aspects due to the coupling of the two order parameters are revealed. To this end we disregard, for the moment, fluctuation effects in Δ because they do not affect qualitatively the new findings which merely depend on the form of the coupling.

As in the standard approach to the theory coupled order parameters, minimization of $\mathcal{F}(\Delta,\Psi)$ with respect to Δ yields

$$|\Delta(\mathbf{r})|^2=-\left[a-\beta^{-2}\sum_n c_n|\Psi_n(\mathbf{r})|^2+g\beta^{-2}\sum_n|\nabla_r\Psi_n(\mathbf{r})|^2\right]\left[b+d\beta^{-4}\sum_{n_i}\Psi^*_{n_1}(\mathbf{r})\Psi_{n_2}(\mathbf{r})\Psi^*_{n_3}(\mathbf{r})\Psi_{n_4}(\mathbf{r})\right]^{-1}. \quad (4)$$

A first interesting point is that the coherence properties of the system lead to a renormalization of $|\Delta|$, usually considered in the literature as a single-grain parameter; this can be of some relevance as will be discussed below.

The effective free energy $\mathcal{F}^{\rm eff}(\Psi)$ is obtained,

$$\mathcal{F}^{\rm eff}(\Psi)=V_\rho^{-1}\int d\mathbf{r}\left\{-a^2/2b+\beta^{-2}\sum_n(1+c_n a/b)\,|\Psi_n(\mathbf{r})|^2 +\beta^{-4}\left[(a^2d/2b^2)\sum_{n_i}\Psi^*_{n_1}(\mathbf{r})\Psi_{n_2}(\mathbf{r})\Psi^*_{n_3}(\mathbf{r})\Psi_{n_4}(\mathbf{r})\delta\left[\sum_{n_i},0\right] -(1/2b)\left[\sum_n c_n\,|\Psi_n(\mathbf{r})|^2\right]^2\right]-\beta^{-2}(ag/b)\sum_n|\nabla_r\Psi_n(\mathbf{r})|^2\right\}. \quad (5)$$

The free energy (4) differs from the one usually treated[1] in the form of the involved coefficients.

An interesting feature clearly revealed by our treatment is the change in the order of the phase-locking transition, this last being a typical occurrence in systems with coupled order parameters.[13] As it is well known, the conditions for the appearance of a tricritical point is the vanishing of both the zero-frequency coefficients of $|\Psi|^2$ and $|\Psi|^4$ in the free-energy expansion

$$1+(a/b)c=0\ , \quad (6)$$

$$da^2/b-c_0^2=0\ . \quad (7)$$

Let us remember that we are dealing with the case $T_J \lesssim T_C$. In this limit we will show that the system exhibits a first-order transition for grain sizes large enough to allow the neglecting of the charging effects. Using the proper expression for the involved coefficients we obtain the following condition for the transition to be first order:

$$\delta<[\pi^2/4(3)]t_J^{-1}(1-t_J)^2\ , \quad (8)$$

where $t_J=T_J/T_C$. It is clear that a lower bound for the grain volume exists in order that the system undergo a second-order phase transition. As shown in Fig. 1 for $t_J \lesssim 1$ a first-order phase transition appears for not-too-small grain volumes as stated above; at lower t_J the transition is of the second order for real systems, so to fall in the framework of the usual phase approximation theory.

It is quite amusing that the appearance of this novel feature in the phase diagram of the system (under proper conditions) streams directly, in a much more realistic context, i.e., accounting for the Δ-Ψ coupling, from a deeper treatment of a model Hamiltonian that is widely accepted—even in its restricted version—as a suitable description of the physics of a granular superconductor.

As discussed in Ref. 12 fluctuations of $|\Delta|$ can modify only quantitatively the obtained phase diagram without infirming the very existence of the newly found tricritical line. It is worth stressing that the renormalization of $|\Delta|$ predicted in Eq. (4) for $T \lesssim T_C$ can be found also in the low-temperature limit.

At lower temperatures, the two order parameters' free energy cannot be expressed in the Ginzburg-Landau form because the usual power expansion in $|\Delta|$ for the single-grain part is not valid, and the coupling is now of the form $|\Delta|\,|\Psi|^2$ as can be readily verified in the proper limit of the well-celebrated Ambegaokar-Baratoff formula. Even if in a more complete treatment the quantum fluctuations in $|\Delta|$ must be taken into account, the static mean-field approximation is enough to show the new findings.

By the same Hubbard-Stratanovich technique applied to Eq. (1) resorting now to the $T\to 0$ limit, it is possible to obtain the appropriate $\mathcal{S}(\Delta,\Psi)$ (we skip the standard calculations for brevity), the minimization of which leads to a generalized BCS equation for Δ. If we are near to the paracoherent transition,

$$2\,|\Delta|\left[-\sum(\zeta^2-E_k^2)^{-1}-\beta\Omega/\lambda\right]+\beta^{-2}\sum_n c_n\,|\Psi_n|^2=0\ , \quad (9)$$

where ζ are the Matsubara frequencies and E_k the quasiparticle excitation energies [the others quantities entering (8) have been already defined]. In Eq. (8) the last term

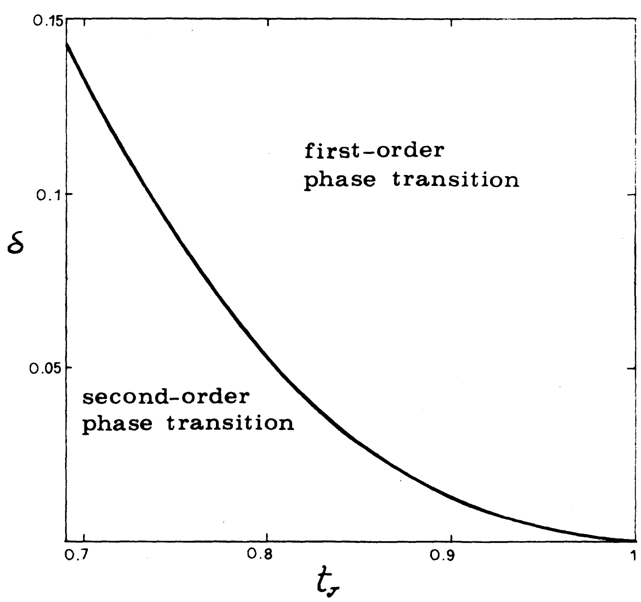

FIG. 1. The phase diagram in the plane t_J-δ showing the curve of the tricritical point; δ is inversely proportional to the volume of the grain and for grain sizes $\simeq 1000$ Å is about 10^{-3} while it approaches unity for sizes of $\simeq 100$ Å. It can be readily observed that the neglecting of charging is consistent with the mentioned values.

represents the correction due to the coherence properties of the array and reduces to the BCS equation when $\Psi=0$. Rearranging Eq. (8), taking the $T\rightarrow 0$ limit, and bearing in mind that near the paracoherent transition Ψ is small, one finds the relative correction for Δ at $T=0$,

$$(\,|\Delta|-|\Delta_0|\,)/|\Delta_0|=(\delta/\beta_c\Delta_0)\bar{c}_0(\alpha-\alpha_{\rm cr})^{2\eta}\ , \quad (10)$$

where Δ_0 is the BCS order parameter of the grain as if alone, $\bar{c}_0=\lim_{\beta\rightarrow\infty}\beta^{-1}w(0)$, $\alpha=zE_J/U$, $\alpha_{\rm cr}$ is the critical value for which $T_J(\alpha)=0$, and η is the critical exponent for Ψ (we did not call it β as usual to avoid a misunderstanding with the inverse of the temperature).

Equation (10) shows several aspects not accounted for in the usual phase Hamiltonian approach: Indeed we predict a scaling law for the depression of the gap as the critical temperature T_J approaches zero [we remind that $T_J\propto(\alpha-\alpha_{\rm cr})^{2\nu}$].

This effect has been detected experimentally by tunneling measurements (see Ref. 14 for a detailed discussion), and it cannot be understood in the framework of the phase model. We admit that we cannot perform a quantitative comparison with the experiments but, in our approach it is a simple consequence of the Δ-Ψ coupling.

It is worth noticing that the "strength" of this correction strongly depends on the prefactor of the rhs of Eq. (9); therefore according to its value this effect could be weak. (This could be the case in the experiments of White *et al.*[15])

In our opinion the scope of this work is twofold and is essentially summarized in Eqs. (5) and (10). First, a "new" free energy is presented for the analysis of the paracoherent transition: it has the usual $O(2)$ symmetry but different coefficients due to Δ-Ψ coupling. The obtained form is far from containing a mere renormalization because as we showed in (7) it leads to the appearance of a tricritical line in the phase diagram. The choice of a mean-field approximation for $|\Delta|$ could be questionable, although it must be stressed that the very existence of the first-order phase transition is not affected by $|\Delta|$ fluctuations[12] and that these last are seriously suppressed because of the coupling between grains.

Second, we show through Eq. (10) how the Δ-Ψ coupling is relevant in the low-temperature region to explain tunneling data. It is our purpose to work out more deeply the consequences of this approach in order to explain other features such as the broadening of the gap edge with increasing R_N which cannot be understood inside the limits we have posed to the actual analysis.

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