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Reentrant phase diagram for granular superconductors

E. Šimánek
Department of Physics, University of California, Riverside, Riverside, California 92521
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The mean-field phase diagram of the self-charging model of a granular superconductor is calculated using the functional-integral formulation. A pronounced reentrance is obtained in quantitative agreement with a previous calculation, involving 2π-antiperiodic eigenstates of the Mathieu equation. It is argued that low-lying, odd-electron-number excitations play a significant role in the thermodynamics of Josephson-junction arrays, for which the Feynman functional-integral formulation holds.

There has been considerable current interest in the problem of phase ordering in granular superconductors taking place in the presence of the electrostatic charging energy.[1-6] The first calculation of the relevant phase diagram has been carried out using the model of a regular three-dimensional array of Josephson junctions with the diagonal form of the charging energy[3] (self-charging model[5]). Treating the Josephson coupling in the mean-field approximation, a phase diagram with well-pronounced reentrant behavior has been obtained.[3] Subsequent analysis of this result has revealed that the reentrance depends on the choice of the interval for the phase ϕ of the superconducting order parameter.[6] In particular, the reentrance disappears[4,7] when this interval is restricted to $-\pi<\phi<\pi$. On the other hand, the reentrant behavior, first obtained in Ref. 3, corresponds to the choice $-\infty<\phi<\infty$. In this case, the mean-field Hamiltonian describes a fictitious particle moving in a $\cos\phi$ potential over the unrestricted ϕ space. By virtue of the Floquet theorem, the periodic eigenfunctions of this problem are not all 2π periodic, but there is a set of 2π-antiperiodic (i.e., 4π-periodic) solutions which have low excitation energy and are directly responsible for the reentrant behavior.[6] One unusual aspect of these 2π-antiperiodic states is that they are associated with odd eigenvalues of the electron number operator ("fractional charges" of the pairs). For this reason, the relevance of such states for the thermodynamics of the phase ordering in granular systems has not been widely accepted.[4,7,8] One exception is the work of Maekawa, Fukuyama, and Kobayashi,[9] in which a reentrant phase transition of the Kosterlitz-Thouless variety has been predicted for two-dimensional arrays upon inclusion of such states. The same problem has recently been studied by José,[10] using Feynman's path-integral formulation of the partition function. The resulting renormalization-group equations, derived in the semiclassical approximation, again indicate the presence of reentrant normal phases, due to the charging energy.

The purpose of the present Rapid Communication is to reexamine the mean-field theory of the self-charging model, starting from a path-integral formulation similar to that of José.[10] It turns out that the resulting phase diagram not only exhibits a pronounced reentrance but also shows a remarkable quantitative agreement with the results of Ref. 3, confirming the importance of the 2π-antiperiodic states. Following the notation of Ref. 3, we consider a three-dimensional regular array of grains whose Hamiltonian is given by

$$H=\frac{U}{2}\sum_i n_i^2+\sum_{\langle ij\rangle} E_J[1-\cos(\phi_i-\phi_j)] \quad , \tag{1}$$

where n_i is the deviation from the average number of electrons on the ith grain. The parameter U is the local charging energy and E_J is the Josephson coupling energy between the nearest-neighbor grains. Introducing the pair number operator $n/2=-i\partial/\partial\phi$, the first term of Eq. (1) becomes the kinetic energy operator of the plane rotator, if ϕ is restricted to $-\pi<\phi<\pi$. Extending this range to $-\infty<\phi<\infty$, makes the operator equivalent to that of the kinetic energy of a free particle moving in the one-dimensional unrestricted ϕ space. Only then the path-integral formulation of Feynman[11] is possible with the partition function of the form[10]

$$Z=e^{-\beta F}=\int_{\text{per}} D\phi(\tau)\exp\left(-\int_0^\beta L(\phi)\,d\tau\right) \quad , \tag{2}$$

where

$$L(\phi)=\frac{1}{8U}\sum_i\left(\frac{\partial\phi_i(\tau)}{\partial\tau}\right)^2+E_J\sum_{\langle ij\rangle}\{1-\cos[\phi_i(\tau)-\phi_j(\tau)]\} \quad . \tag{3}$$

The subscript on the path integral (2) indicates that only those paths are to be included, for which the periodic boundary condition $\phi_i(0)=\phi_i(\beta)$ holds. It is important to point out that the expressions (2) and (3) can also be derived from a microscopic Hamiltonian of the array by means of the Hubbard-Stratonovich transformation,[12] as recently applied to the single Josephson junction by Ambegaokar, Eckern, and Schön.[13] To calculate the transition temperature of the array in the mean-field approximation, we start from the variational principle for the free energy[11]

$$F\le F_0+\frac{1}{\beta Z_0}\int_{\text{per}} D\phi\exp(-S_0[\phi])(S-S_0)=F_t \quad , \tag{4}$$

with $S=\int_0^\beta L(\phi)\,d\tau$ and

$$e^{-\beta F_0}=Z_0=\int_{\text{per}} D\phi\exp(-S_0[\phi]) \quad . \tag{5}$$

The trial action $S_0=\int_0^\beta L_0\,d\tau$ involves the mean-field choice for L_0 of the form

$$L_0=\sum_i\left[\frac{1}{8U}\left(\frac{\partial\phi_i}{\partial\tau}\right)^2+z\gamma E_J\cos\phi_i(\tau)\right] \quad , \tag{6}$$

where z is the coordination number in the array. The variational parameter γ is determined from Eq. (4) by requiring that $\delta F_t=0$ when $\gamma \to \gamma+\delta\gamma$. This yields a self-consistent equation for γ:

$$\gamma=\frac{2}{Z_0}\int_{\text{per}} D\phi \exp(-S_0[\phi])\cos\phi_j=2\langle\cos\phi_j\rangle_0 \ . \tag{7}$$

Near the transition temperature $T_c=\beta_c^{-1}$, the parameter γ is small and one can use the expansion of S_0 in evaluating the path integral (7)

$$\langle\cos\phi_j(\tau)\rangle_0 \cong \frac{1}{Z_U}\int_{\text{per}} D\phi \exp(-S_U[\phi])\left[1+z\gamma E_J\sum_i\int_0^{\beta_c} d\tau' \cos\phi_i(\tau')\right]\cos\phi_j(\tau) \ , \tag{8}$$

where

$$S_U=\int_0^{\beta_c} d\tau\left[\frac{1}{8U}\sum_i\left(\frac{\partial\phi_i}{\partial\tau}\right)^2\right] \tag{9}$$

and

$$Z_U=\int_{\text{per}} D\phi \exp(-\beta S_U[\phi]) \ . \tag{10}$$

Since only the $i=j$ term contributes in Eq. (8), we obtain, taking into account the equivalence of all sites

$$\langle\cos\phi_j(\tau)\rangle=\langle\cos\phi(\tau)\rangle_0$$

$$\cong zE_J\gamma\int_0^{\beta_c} d\tau'\langle\cos\phi(\tau')\cos\phi(\tau)\rangle_U \ , \tag{11}$$

where the averaging in the phase correlator in Eq. (11) is to be done with use of the action (9). Introducing γ from Eq. (7), we obtain from Eq. (11) the equation for the transition temperature

$$1=2zE_J\int_0^{\beta_c} d\tau\langle\cos\phi(\tau)\cos\phi(0)\rangle_U \ . \tag{12}$$

This equation agrees with the self-consistency equation for T_c derived previously by Efetof.[4] The present approach differs from Efetof's in the method of the evaluation of the phase correlator. We use the path-integral approach of Feynman,[11] in which $\phi(\tau)$ is a classical variable, so that we have

$$\langle\cos\phi(\tau)\cos\phi(0)\rangle_U=\tfrac{1}{2}\langle\exp\{i[\phi(\tau)-\phi(0)]\}\rangle_U$$

$$=\tfrac{1}{2}R(\tau) \ , \tag{13}$$

where, according to Eqs. (8)–(11)

$$R(\tau)=\frac{1}{Z_U}\int_{\text{per}} D\phi(\tau)\exp(-S_U[\phi])\exp\{i[\phi(\tau)-\phi(0)]\} \ . \tag{14}$$

To satisfy the periodicity requirement $\phi(0)=\phi(\beta)$, we expand $\phi(\tau)$ into a Fourier series[11]

$$\phi(\tau)=\phi(0)+\sum_{n=1}^{\infty}\phi_n\sin\frac{n\pi\tau}{\beta_c} \ . \tag{15}$$

Introducing Eq. (15) into Eq. (14), we have

$$R(\tau)=\frac{\int\int\int_{-\infty}^{\infty}\exp\left[-\sum_{n=1}^{\infty}(A_n\phi_n^2-B_n\phi_n)\right]d\phi_1 d\phi_2\cdots}{\int\int\int_{-\infty}^{\infty}\exp\left[-\sum_{n=1}^{\infty}A_n\phi_n^2\right]d\phi_1 d\phi_2\cdots} \ , \tag{16}$$

where

$$A_n=\frac{n^2\pi^2}{16U\beta_c}, \quad B_n=i\sin\frac{n\pi\tau}{\beta_c} \ .$$

Evaluating the multiple Gaussian integrals, we obtain from Eq. (16)

$$R(\tau)=\exp\left[\sum_{n=1}^{\infty}\frac{B_n^2}{4A_n}\right]$$

$$=\exp\left[-\frac{4U}{\beta_c}\sum_{n=1}^{\infty}\frac{\sin^2(n\pi\tau/\beta_c)}{(n\pi/\beta_c)^2}\right]$$

$$=\exp[-(2U/\beta_c)\tau(\beta_c-\tau)] \ . \tag{17}$$

In comparison with the phase correlator of Efetof,[4] this expression is extremely simple. The summation over the eigenstates of the Coulomb Hamiltonian does not explicitly enter in $R(\tau)$, and hence the numerical calculation of the transition temperature T_c is considerably simplified. Using Eqs. (13) and (17) in Eq. (12), we obtain the self-consistent equation for T_c in the form

$$1=\left(\frac{zE_J}{T_c}\right)\frac{y(x)}{x} \ , \tag{18}$$

where $x=\sqrt{U/2T_c}$ and $y(x)$ is the Dawson integral:[14]

$$y(x)=\exp(-x^2)\int_0^x\exp(t^2)\,dt \ . \tag{19}$$

Following Ref. 3, we introduce the ratio $\alpha=zE_J/U$ and the transition temperature T_c^c of the classical model ($U=0$), which turns out equal to zE_J. Then Eq. (18) can be written

$$1=2\alpha xy(x)=2\alpha P(x) \ , \tag{20}$$

which is to be solved numerically for x. The values of x, thus obtained, determine the transition temperature from the relation

$$\frac{T_c}{T_c^c}=\frac{1}{2\alpha x^2} \ . \tag{21}$$

The resulting phase diagram is shown in Fig. 1. It exhibits a reentrant bulge protruding over the region $0.78<\alpha<1$. The inset illustrates the numerical solution of Eq. (20). It is clearly the nonmonotonic behavior of the function $P(x)$ which is responsible for the reentrance. The maximum of $P(x)=xy(x)$ coincides with the inflection point of $y(x)$, which is known with high accuracy.[14] Thus we obtain $P(x_{\max})=0.64$, which exceeds the asymptotic value $P(x\to\infty)=0.5$. The double-valued solutions for x are obtained when the parameter $1/2\alpha$ is confined between these two values (lines a and b). The numerical values of $\alpha_{\min}=0.78$ $(T_c/T_c^c)_{\alpha_{\min}}=0.29$ agree remarkably well with the results of Ref. 3. This agreement is not entirely unexpected, since the approach of Ref. 3, based on averaging over the eigenstates of Mathieu's equation, and the Feynman path-integral approach are both dealing with the same

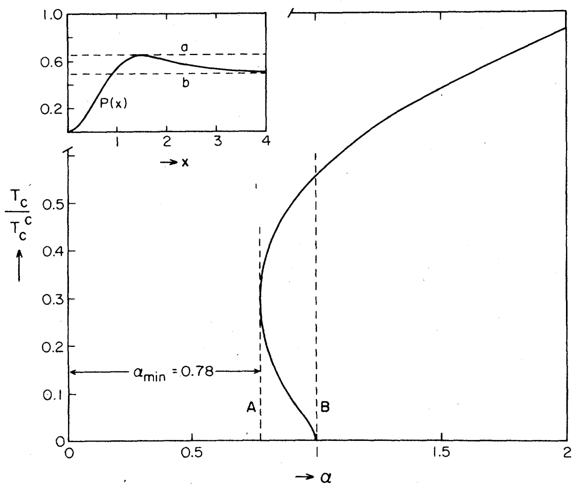

FIG. 1. Phase-ordering temperature ratio T_c/T_c^c plotted as a function of parameter $\alpha=zE_J/U$ from Eqs. (20) and (21). Inset: Plot of function $P(x)=xy(x)$ based on Ref. 14. The dashed lines a and b indicate the limits between which the numerical solutions of Eq. (20) produce the reentrant portion of the phase diagram, confined between the dashed lines A and B.

problem: i.e., a fictitious particle moving in a $\cos\phi$ potential, defined over $-\infty<\phi<\infty$. It should be pointed out that the partition function of a free plane rotator, corresponding to the choice $-\pi<\phi<\pi$, is not given by Eqs. (9) and (10), but by the Villain form of $\cos\phi$ interaction along the τ axis.[5,15] The latter involves Fourier components $\exp(im\phi)$ with integer quantum numbers m only. In contrast, the path integral (10) corresponds to a free particle Hamiltonian with a continuous translational symmetry in the ϕ space, and hence it includes all paths with a continuous spectrum of quantum numbers m. The symmetry is reduced to the discrete translations after the $\cos\phi$ potential is introduced and the representations of the corresponding discrete group involve both the 2π-periodic and 2π-antiperiodic states. The fact that the latter states are contained in the Feynman path integral (10) helps us understand the agreement of the present calculation with the results of Ref. 3.

As previously pointed out,[6] the phase-locking transition in a granular superconductor is not necessarily isomorphous to that in the x-y model, in which the angular variable is confined to the interval $(-\pi,\pi)$. This is clearly demonstrated, for instance, in the "tight-binding" approach to the Josephson effect by Ferrell and Prange.[16] Any real value of ϕ is allowed in the expression for the Josephson coupling energy, derived by these authors (see also Ref. 17). The preference for the Feynman form of the charging energy is further substantiated by the microscopic derivation of the path-integral formulation for Josephson junctions with charging energy.[13] As pointed out in Ref. 13, the bulk energies of the superconductors tend to pin $\dot\phi_i$ to the local potential V_i. It turns out that for the self-charging model the corresponding Josephson relation $\dot\phi_i=2\,eV_i$ directly leads to the Feynman form of the action, given by Eq. (3) with no restriction on the range of ϕ_i.

In conclusion, the functional-integral formulation for the array of Josephson junctions confirms the reentrant feature of the mean-field phase diagram, previously attributed to the thermally excited 2π-antiperiodic eigenstates of the Mathieu's equation.[3] This suggests that the excitations have a real physical significance in the thermodynamics of Josephson-junction arrays. It should be interesting to apply the functional-integral approach to a more realistic model, involving the off-diagonal (grain-grain) Coulomb interaction.[4,5,8] The significance of such efforts seems underlined by the recent experimental observation of a reentrant superconducting resistive transition in granular $BaPb_{0.75}Bi_{0.25}O_3$ superconductor.[18]

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