Expansion in $1/z$ for the Transition Temperature of Granular Superconductors

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An expansion in powers of $1/z$ is used to study the effect of phase fluctuations on the transition temperature of a granular superconductor, modeled as an array of Josephson junctions. The lowest-order correction to T_c/zJ diverges to negative infinity at the grain diameter $\alpha = zJ/U = 2$ and reaches the value $-1/2z$ when $\alpha = \infty$, which corresponds to the classical XY model. The lowest-order correction to the critical grain diameter is also calculated.

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It is common in condensed matter physics to use approximations that introduce unknown error into physical solutions. Mean-field (MF) and self-consistent harmonic (SCH) methods dominate the study of granular superconductors, yet the errors introduced by these approximations are not well understood. While the SCH approximation violates the phase periodicity of the Josephson energy, the MF theory neglects the coupling of phase fluctuations on neighboring grains. Both approximations break down when phase fluctuations become large, such as near the transition temperature T_c. The effects of phase fluctuations in shifting T_c from its MF or SCH value has been largely unknown.

In this Letter, I describe a new technique for systematically studying the effects of phase fluctuations in granular superconductors by performing an expansion in $1/z$, where z is the number of nearest neighbors for each grain. The zeroth-order term in the expansion of any thermodynamic quantity is the MF value. The first-order correction involves the coupling of phase fluctuations neglected by MF theory. To demonstrate this approach, I calculate the first-order correction to the transition temperature of a granular superconductor. I find that fluctuations decrease the transition temperature and increase the critical grain diameter below which superconductivity becomes impossible.

It is customary to model a granular superconductor by an array of Josephson-coupled grains. The Hamiltonian for such an array is

$$H = 2U\sum_i n_i^2 + J\sum_{\langle i,j\rangle}[1-\cos(\phi_i-\phi_j)]\,, \tag{1}$$

where J is proportional to the probability of Copper-pair tunneling between neighboring grains, $U=e^2/C$ is inversely proportional to the capacitance C of a grain,[1] ϕ_i is the phase of the superconducting order parameter, and $n_i = -id/d\phi_i$ is the operator for the number of excess Cooper pairs on the ith grain. The tunneling of Cooper pairs between neighboring grains favors the growth of a global order parameter with all the phases equal. But the transfer of Cooper pairs between grains costs the charging energy $2U\sum_i n_i^2 = \sum_i q_i^2/2C$, where $q_i = 2en_i$ is the excess charge on a grain. Hence, the charging energy inhibits the tunneling of Cooper pairs and disrupts the phase coherence of the array. The resistivity of a granular superconductor vanishes upon the onset of global phase coherence, below the transition temperature T_c, when $M \equiv \langle\cos\phi_i\rangle$ becomes nonzero. As the grain diameter decreases, the charging energy dominates the Josephson energy and T_c is suppressed. The magnitude of this suppression is measured by the dimensionless parameter $\alpha = zJ/U$, which is proportional to the grain diameter. When $\alpha = zJ/U$ is lower than a critical value α_c, superconductivity becomes impossible and $T_c = 0$.

The Hamiltonian of Eq. (1) has been studied by a variety of analytic methods.[2-11] MF[3-5] and SCH[6] methods agree that the order parameter M is a monotonically decreasing function of temperature and that T_c is a monotonically increasing function of α, for $\alpha > \alpha_c$. However, both analytic techniques break down when phase fluctuations become large. The SCH approximation violates the phase periodicity of the Hamiltonian, $H(\phi_i + 2\pi) = H(\phi_i)$, by replacing the Josephson energy by a quadratic potential. The difference between the quadratic and cosine potentials become important when $\langle(\phi_i-\phi_j)^2\rangle \gtrsim (\pi/4)^2$. Then the phase periodicity becomes crucial[12] and the SCH method becomes inadequate. Because the harmonic eigenfunctions are themselves nonperiodic, it is impossible to restore the effect of phase periodicity in a correction to the lowest-order SCH result. MF theory, on the other hand, maintains the phase periodicity of the array but neglects the coupling of phase fluctuations on neighboring grains. Near the transition temperature T_c, the coupling of phase fluctuations becomes important and the MF theory becomes invalid. Fortunately, the effect of phase fluctuations can be included in a first-order $1/z$ correction to the MF result.

The expansion of the order parameter M in powers of $1/z$ was summarized in previous work[13] and will be described in detail elsewhere.[14] Here I briefly outline the procedure, which involves separating the Hamiltonian

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into three terms:

$$H = H_{\text{eff}} + H_1 + H_2 , \tag{2}$$

where

$$H_{\text{eff}} = \sum_i \{2Un_i^2 - zJ\langle\cos\phi_1\rangle_{\text{MF}}\cos\phi_i\} , \tag{3}$$

$$H_2 = -J\sum_{\langle i,j\rangle} \{(\cos\phi_i - \langle\cos\phi_1\rangle_{\text{MF}})(\cos\phi_j - \langle\cos\phi_1\rangle_{\text{MF}}) + \sin\phi_i\sin\phi_j\} , \tag{4}$$

and H_1 is a c number. The MF theory neglects H_2, which couples the phase fluctuations on neighboring grains.

In the interaction representation, the order parameter is given[15] by

$$M \equiv \langle\cos\phi_1\rangle = \frac{1}{Z}\text{Tr}\left[e^{-\beta H_{\text{eff}}}T_\tau \exp\left(-\int_0^\beta \hat{H}_2(\tau)d\tau\right)\cos\hat{\phi}_1(0)\right] , \tag{5}$$

$$Z = \text{Tr}\left[e^{-\beta H_{\text{eff}}}T_\tau \exp\left(-\int_0^\beta \hat{H}_2(\tau)d\tau\right)\right] , \tag{6}$$

where $\beta = 1/T$, T_τ is the time-ordering operator, and operators in the interaction representation are defined by

$$\hat{A}(\tau) = e^{\tau H_{\text{eff}}}Ae^{-\tau H_{\text{eff}}} . \tag{7}$$

The $1/z$ expansion is generated from Eqs. (5) and (6) by expanding both the numerator of M and the partition function Z in powers of fluctuation energy H_2. Expressed as functions of the dimensionless temperature $T^* = T/zJ$ and the grain diameter α, the terms in this expansion can be classified by their order in $1/z$.

To first order in $1/z$, M is written

$$M = M_0(\alpha, T^*) + \frac{1}{z}M_1(\alpha, T^*) . \tag{8}$$

The lowest-order term M_0 is the MF solution, which is a function only of α and T^*. The first-order correction involves a sum over an infinite number of terms, shown in Fig. 1. Each line represents a factor of H_2 coupling neighboring grains. The origin of every diagram is fixed at grain 1 but the other points are free to vary. Since each link (made up of a single line or a loop of two lines) can be oriented in z different directions, the contribution of a diagram with m lines is proportional to $J^m z^{m-1} = (Jz)^m/z$. Thus, in terms of the dimensionless variables, the mth-order diagram is of order $1/z$.

In two or three dimensions, the contribution of closed paths to M_1 can be neglected. The number of closed-path diagrams with m lines is almost always[16] less than z^{m-2}, so the contribution of such diagrams is of order $1/z^2$ or lower. Since closed-path diagrams involve the connectivity of the array, their contribution will depend on the lattice as well as on the number of nearest neighbors. The correction M_1/z, on the other hand, is the same for a hexagonal lattice in two dimensions and a cubic lattice in three dimensions, both with $z = 6$. Hence, the first-order correction M_1/z can be considered the lattice-independent correction to the order parameter.

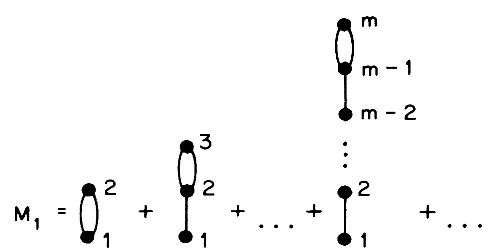

FIG. 1. Series of diagrams which contribute to M_1.

In Ref. 13 the infinite sum for M_1 was performed by expressing the $(m+1)$th-order contribution $M_1^{(m+1)}$ in terms of the mth order one $M_1^{(m)}$. Because $M_1^{(m+1)} = fM_1^{(m)}$, the summation $M_1 = \sum_{m=2} M_1^{(m)} = M_1^{(2)}/(1-f)$ can be evaluated exactly. The first-order correction M_1 is negative and increases in magnitude as T^* increases or as α decreases, so that fluctuations suppress the order parameter. As T^* approaches the MF transition temperature, the scaling function f reaches 1 and M_1 diverges to $-\infty$. This divergence signals the breakdown of the expansion about the MF solution near the transition temperature, when fluctuations drive the system normal.

The divergence of M_1 at the MF transition temperature also signals a shift in the transition temperature away from the MF value. This shift can be calculated by using

$$T_c^* = T_0(\alpha) + \frac{1}{z}T_1(\alpha) \tag{9}$$

to satisfy the condition $M(T_c^*) = 0$. The first-order correction T_1 obtained from Eqs. (8) and (9) is

$$T_1(\alpha) = -\lim_{T^*\to T_0} \frac{M_1(\alpha, T^*)}{dM_0/dT^*} . \tag{10}$$

Both the MF transition temperature T_0 and the first-order correction T_1 are plotted in Fig. 2 versus grain diameter α. The MF transition temperature T_0 is suppressed by charging effects and vanishes when the grain diameter α is less than 2. As expected, the fluctuations neglected by MF theory decrease the transition tempera-

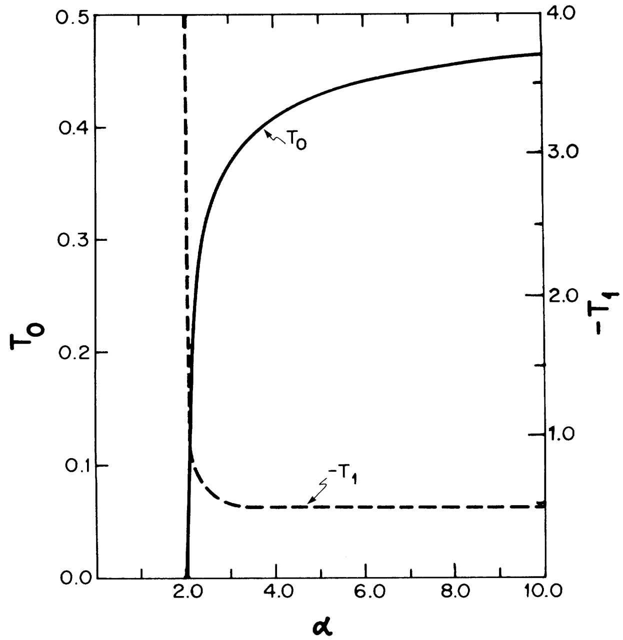

FIG. 2. T_0 (solid) and $-T_1$ (dashed) vs α.

ture: The first-order correction T_1 is negative and increases in magnitude as α decreases. In the limit $\alpha \rightarrow \infty$, corresponding to a classical XY Hamiltonian without charging energy, $T_0 \rightarrow \frac{1}{2}$ and $T_1 \rightarrow -\frac{1}{2}$. The deviation from this limit in Fig. 2 arises from computational uncertainty. On the other hand, when $\alpha \rightarrow 2$, T_1 diverges to $-\infty$.

Just as the divergence in M_1 signals a shift in T_c^*, the divergence in T_1 likewise signals a shift in the critical grain diameter α_c, below which superconductivity is impossible. Writing

$$\alpha_c = \alpha_0 + \frac{1}{z}\alpha_1 , \tag{11}$$

the first-order correction to the MF value $\alpha_0 = 2$ is given by

$$\alpha_1 = -\lim_{\alpha \rightarrow \alpha_0} \frac{T_1(\alpha)}{dT_0/d\alpha} = \frac{7}{5} . \tag{12}$$

Thus, fluctuations increase the critical grain diameter and eliminate superconductivity for grains with diameters α between 2 and $2+7/5z$. Using a cluster-expansion method at zero temperature, Ferrell and Mirhashem[11] also obtained the result $\alpha_1 = \frac{7}{5}$. Because their method is very different from the technique described here, the agreement between the two approaches is quite gratifying. The detailed calculation required to obtain the result of Eq. (12) will be described elsewhere.[14]

Quantitatively, the calculations of this work indicate that fluctuations induce significant shifts in both the transition temperature and the critical grain diameter. For a cubic lattice with $z=6$, the shift in T_c^* for large grains is about 15%. For smaller grains, this shift becomes even larger. It would be very interesting to verify the predictions of this work for $\alpha = \infty$ by comparing the transition temperatures of two-dimensional square ($z=4$) and hexagonal ($z=6$) arrays. For $\alpha = \infty$,

$$T_c^* = \frac{1}{2}\left(1 - \frac{1}{z}\right) , \tag{13}$$

so that to first order, $T_c(z=4) = 3T_c(z=6)/5$.

Strictly speaking, of course, an expansion in powers of $1/z$ is invalid in two dimensions, where spin waves destroy the long-range order. In two dimensions, the MF values of the order parameter and the transition temperature are exactly canceled by the fluctuation terms of the expansion, summed to infinite order in $1/z$. This cancellation does not appear at any finite order in the $1/z$ expansion because the MF theory has replaced the low-lying spin waves by a collection of Einstein modes with discrete frequencies. The spin-wave spectrum is fully recovered and the phase coherence is completely destroyed only after all the diagrams have been summed.

To conclude, I have demonstrated a new approach to calculate the lattice-independent correction to the MF transition temperature and critical grain diameter of a granular superconductor. Previously, $1/z$ expansions were used to study the Heisenberg and Ising models in a magnetic field.[17] However, these expansions were rather unwieldy because an infinite summation was required to obtain the lowest-order MF results. The $1/z$ expansion developed in this work avoids that task by expanding about the MF Hamiltonian. This method has recently[14] been used to study the short-range order and specific-heat anomalies of granular superconductors.

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[16]An exception occurs for close-packed arrays in three dimensions, when $z=12$ and the number of closed paths with three lines is $24>z$. However, the z^2 "open" diagrams still greatly outnumber the closed-path diagrams with three lines and the number of closed paths with $m>3$ lines *is* less than z^{m-2}.

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