PHYSICAL REVIEW B VOLUME 37, NUMBER 1 1 JANUARY 1988

Universal criterion for the onset of superconductivity in granular films

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(Received 19 August 1987)

In a granular film, modeled by a Josephson-coupled lattice, virtual tunneling of quasiparticles results in a critical value $R_\square^c$ for the normal-state sheet resistance. $R_\square^c$ is a universal upper bound for zero-temperature superconductivity and is computed in descending powers of z, the number of grain neighbors. Mean-field theory for a square lattice gives the leading term $R_\square^c = 5.7$ kΩ. For the fractional correction from the next term in the expansion, the Bethe approximation gives $-3/5z^2$, or -4% for $z=4$.

Ever since the discovery of quantum mechanics, its applicability to macroscopic systems has been a subject of great interest. The recently reported universal criterion[1] for the onset of superconductivity in granular films, we assert, serves to confirm the validity of quantum mechanics in describing the collective behavior of the grains, each of which contains on the order of 10^4–10^6 electrons. In this Rapid Communication we put forward a simple quantum-mechanical treatment of the collective effect of the Josephson coupling between neighboring grains in a granular film. Our computation predicts a threshold resistance per square of approximately 5.7 kΩ, above which the film will not be superconducting. In view of the various approximations on which this prediction is based, we think that it is in satisfactory accord with the reported[1] empirical critical resistance of $R_\square^c = 6.5$ kΩ.

Our study is limited to the ground state of the granular film at a temperature $T=0$. The "macroscopic" collective variables are ϕ_i, where ϕ_i is the phase of the Bardeen-Cooper-Schrieffer (BCS) ground-state wave function within the ith grain. The "velocities" are given by the usual quantum-mechanical expression

$$\dot{\phi}_i = \frac{2e}{\hbar} V_i \ , \tag{1}$$

where $-e$, $2\pi\hbar = h$, and V_i are the electron charge, Planck's constant, and the electrostatic potential of the ith grain, respectively. Substituting Eq. (1) into the total electrostatic energy of the array of grains yields the "kinetic energy"

$$\begin{aligned} W &= \tfrac{1}{2}\sum_i C_i V_i^2 + \tfrac{1}{2}\sum_{ij} \Delta C_{ij}(V_i - V_j)^2 \\ &= \frac{\hbar^2}{8e^2}\left[\sum_i C_i \dot{\phi}_i^2 + \sum_{ij} \Delta C_{ij}(\dot{\phi}_i - \dot{\phi}_j)^2\right] \ , \end{aligned} \tag{2}$$

where C_i is the capacitance of the ith grain to ground and ΔC_{ij} is the additional mutual capacitance between neighboring grains. The double sum is taken over all neighboring pairs. The total energy of Josephson coupling between neighboring pairs is

$$U = -\sum_{ij} E_J^{ij} \cos(\phi_i - \phi_j) \ . \tag{3}$$

Denoting the charge on the ith grain by Q_i, we find that the total Josephson current flowing into the ith grain from its neighbors is

$$\begin{aligned} \dot{Q}_i &= \frac{-2e}{\hbar}\frac{\partial U}{\partial \phi_i} \\ &= C_i \dot{V}_i + \sum_j \Delta C_{ij}(\dot{V}_i - \dot{V}_j) \\ &= \frac{\hbar}{2e}\left[C_i \ddot{\phi}_i + \sum_j \Delta C_{ij}(\ddot{\phi}_i - \ddot{\phi}_j)\right] \\ &= \frac{2e}{\hbar}\frac{d}{dt}\frac{\partial W}{\partial \dot{\phi}_i} \ , \end{aligned} \tag{4}$$

the second line corresponding to the definition of capacitance as a linear response function. The sum is over the neighbors of the ith grain. The third and fourth lines follow from the substitution of Eqs. (1) and (2), respectively. Comparison of the first line of Eq. (4) and the fourth line reveals that it is nothing other than the Euler-Lagrange equation based on the Lagrangian $L = W - U$.

Passing from L to the corresponding Hamiltonian leads to a complicated nonlinear quantum-mechanical many-body problem. Some aspects of this problem, particularly its long-wavelength behavior, have been discussed by Chakravarty, Kivelson, Zimanyi, and Halperin.[2] The occurrence of Goldstone modes in the limiting case that we will study can be expected to destroy long-range phase coherence without necessarily preventing the superconductivity of the film. Comparison with one-dimensional superconductivity is useful in this context. Although fluctuations do destroy the long-range correlation of the phase in such a system,[3] it has been noted[4] that resistance is generated only by topology-changing fluctuations of the tunneling type, which become exponentially weak as $T \to 0$. In this note, we concentrate on the short-range features of the problem, which are less affected by the fluctuations. We seek a criterion for superconductivity of the granular film in terms of descending powers of z, the number of neighbors of a gain. In the limit $z \to \infty$, the fluctuations average out and the mean-field treatment becomes exact. In studying the problem as a function of z, we adopt an approach somewhat different from our previous mean-field treatment[5] and from the earlier mean-field treatments of Simánek,[6] Doniach,[7] Efetov,[8] and Fazekas, Mühlschlegel, and Schröter.[9] The latter half of this paper is devoted to estimating finite z corrections by means of the Bethe approximation and some generalization of it. These $z<\infty$ corrections will be seen to be relatively small, indicating that mean-field theory may be a satisfactory approximation for determining the onset criterion.

The configuration of the ith grain is conveniently specified by the phase factor $\exp(i\phi_i)$, whose ground-state expectation value can be written as

$$\langle e^{i\phi_i}\rangle = \mu_i e^{i\alpha_i} \; . \tag{5}$$

At this point, we neglect the disorder in the granular film. This permits us to drop the subscript on the order parameter μ. Furthermore, since the phase fluctuations are primarily due to the long-wavelength Goldstone modes, we assume that $\alpha_j \simeq \alpha_i$ is practically constant in the vicinity of the ith grain and can therefore be gauged to zero. Thus, we have $\langle \sin\phi_i\rangle = 0$ and $\langle\cos\phi_i\rangle = \mu$, so that the intergrain Josephson interaction acting on the ith grain as a consequence of the average over the jth neighbor is

$$\langle\cos(\phi_i-\phi_j)\rangle = \cos\phi_i\langle\cos\phi_j\rangle + \sin\phi_i\langle\sin\phi_j\rangle = \mu\cos\phi_i \; . \tag{6}$$

The assumption of a perfect lattice enables us to drop the subscripts and superscripts on the parameters of L. Moreover, in studying the response of the ith grain to the mean-field set up by the jth neighbor, we can regard the latter as static and neglect its "velocity" $\dot\phi_j$. Therefore, the effective single-grain Lagrangian is

$$L_1 = \frac{\hbar^2}{8e^2}(C+z\Delta C)\dot\phi_i^2 + z\mu E_J\cos\phi_i \; . \tag{7}$$

The dimensionless conjugate momentum is

$$p_i = \frac{1}{\hbar}\frac{\partial L_1}{\partial\dot\phi_i} = \frac{\hbar}{4e^2}(C+z\Delta C)\dot\phi_i \; . \tag{8}$$

The corresponding Hamiltonian is

$$H_1 = \frac{4e^2}{C+z\Delta C}\left(\frac{p_i^2}{2} - g\cos\phi_i\right) \equiv \frac{4e^2}{C+z\Delta C}\tilde H_1 \; , \tag{9}$$

with the reduced dimensionless Hamiltonian $\tilde H_1$, contained within the parentheses, depending only on the single parameter

$$g = z\left(z+\frac{C}{\Delta C}\right)\mu g_0 \; , \tag{10}$$

where

$$g_0 = \frac{\Delta C E_J}{4e^2} \; . \tag{11}$$

We have found it possible to compute the ground-state energy $E_G(g)$ from a simple variational calculation.[10] The resulting values are in good agreement with the tabulated solution[11] of Mathieu's equation for all values of g. For present purposes, however, we require $E_G(g)$ only for $0<g\ll 1$ at the onset of ordering. In this range, a Taylor's expansion in powers of g is available,[11] from which we obtain

$$\langle\cos\phi_i\rangle = -\frac{dE_g}{dg} = 2g - 7g^3 + \tfrac{116}{3}g^5 - \frac{68687}{288}g^7 + \cdots \le 2g \; , \tag{12}$$

the inequality following from the convexity theorem.[12] Self-consistency requires that we identify the left-hand member of Eq. (12) with μ, the order parameter. Substituting Eq. (10) into the right-hand member of Eq. (12) and canceling μ from both sides yields

$$g_0 \ge 1 \Big/ 2z\left(z+\frac{C}{\Delta C}\right) \; . \tag{13}$$

For very small grains, C can be neglected compared to ΔC, so that we arrive at the universal superconducting phase transition criterion

$$g_0^c = \frac{1}{2z^2} \; . \tag{14}$$

The criterion of Eq. (14) can be expressed in terms of the parameters of the tunneling junction by virtue of the virtual tunneling of quasiparticles,[13,14] which gives rise to ΔC. The frequency-dependent function $\Delta C(\omega)$ can be conveniently calculated from the quasiparticle excitation spectrum by means of the Kramers-Kronig relations.[15] Its zero-frequency limit $\Delta C(0)$ can be expressed in terms of the admittance,

$$\omega_{\rm BCS}\Delta C(0) = \frac{3\pi}{16}\sigma_N \; , \tag{15}$$

where σ_N is the normal-state conductance of the junction. For dimensional reasons, we have introduced the BCS gap frequency $\omega_{\rm BCS} = 2\Delta/\hbar$, where Δ is the BCS energy gap. Neglecting both the phase and frequency dependence of ΔC, it follows[13,14] that

$$\Delta C = \frac{3}{64}\frac{h}{\Delta}\sigma_N \; . \tag{16}$$

Substituting Eq. (16) and the standard expression

$$E_J = \frac{h\Delta}{8e^2}\sigma_N \tag{17}$$

into Eq. (11) yields

$$g_0 = \frac{3}{128}\left(\frac{h\sigma_N}{4e^2}\right)^2 = \frac{3}{128}r^{-2} \; , \tag{18}$$

where the normal-state resistance, measured in units of $R_0 \equiv h/4e^2 = 6.5\ \mathrm{k\Omega}$, is

$$r = \frac{\sigma_N^{-1}}{h/4e^2} = \frac{R_N}{R_0} \; . \tag{19}$$

Substituting Eq. (14) into Eq. (18) gives the universal critical resistance, above which the superconductivity disappears:

$$r_c = \frac{\sqrt{3}}{8}z \; . \tag{20}$$

For a regular square lattice, $z=4$, and Eq. (20) becomes

$r_c = \frac{1}{2}\sqrt{3} = 0.87$, corresponding to a resistance per square of

$$R^c_\square = R^c_N = 5.7\ \mathrm{k\Omega}\ . \tag{21}$$

For a regular hexagonal lattice, $z = 6$, and Eq. (20) yields $r_c = 3\sqrt{3}/4 = 1.3$, corresponding to a resistance per square of $R^c_\square = R^c_N/\sqrt{3} = 4.9\ \mathrm{k\Omega}$. Because of the approximations involved in the calculation (neglect of frequency dependence and of disorder), either of the numerical results can be considered to be in satisfactory agreement with the empirical universal threshold resistance of $R^{\mathrm{expt}}_\square = 6.5\ \mathrm{k\Omega}$.

We now turn to the question of the accuracy of the mean-field result, Eq. (14), by seeking to develop a Laurent series for g^c_0 in descending powers of z. The first step in improving upon the mean-field treatment is to single out two neighboring grains i and j for special attention—namely, the Bethe approximation. These two grains are each acted upon by the mean fields of their $z-1$ neighbors, so that the effective two-grain Lagrangian is

$$L_2 = \frac{\hbar^2 \Delta C}{4e_2}\left[\frac{z}{2}\dot\phi_i^2 + \frac{z}{2}\dot\phi_j^2 - \dot\phi_i\dot\phi_j\right] + (z-1)\mu E_J H' + E_J H''\ , \tag{22}$$

where

$$H' = \cos\phi_i + \cos\phi_j \tag{23a}$$

and

$$H'' = \cos(\phi_i - \phi_j)\ . \tag{23b}$$

Upon inversion of the quadratic-form matrix, the negative cross term in the velocities becomes a positive cross product in the momenta. We find, for the effective two-grain Hamiltonian,

$$H_2 = \frac{4e^2 z}{\Delta C(z^2-1)}\tilde H_2\ , \tag{24}$$

where the reduced Hamiltonian is

$$\tilde H_2 = \tfrac{1}{2}(p_i^2 + p_j^2) + \frac{1}{z}p_i p_j - g_1 H' - g_2 H''\ . \tag{25}$$

The single-grain and two-grain coupling constants are

$$g_1 = (z-1)\mu g_2 \tag{26a}$$

and

$$g_2 = (z - z^{-1})g_0\ . \tag{26b}$$

As before, we set C equal to zero and continue to neglect the frequency dependence and the disorder. The ground-state energy $E_G(g_1,g_2)$ is now a function of both coupling constants. But because we are interested here only in the threshold, where $\mu \to 0$, we need E_G only to second order in g_1. Furthermore, from Eqs. (14) and (26b), we expect g_2 to be of order z^{-1}, so a second-order computation also in g_2 will suffice. In other words, we need to apply third- and fourth-order perturbation theory to the perturbing term in Eq. (25), $-g_1H' - g_2H''$.

Figure 1 exhibits typical Feynman graphs for the Bethe approximation to the desired order in perturbation theory. The interaction of the two grains (solid lines) is indicated by the dashed lines, while the crosses represent the action of the mean field of the $z-1$ neighbors. To third order, i.e., to $O(g_1^2 g_2)$, there is no correction to the mean-field formula, Eq. (14). This becomes evident from combining graphs (a) and (b) of Fig. 1, thereby generating the mean field of the upper grain acting on the lower grain. The mean field is then brought up to its full strength corresponding to z neighbors. As a consequence, the first nonvanishing correction term in Eq. (14) is of $O(z^{-4})$. We get an estimate of the numerical coefficient by going to fourth order, i.e., to $O(g_1^2 g_2^2)$ and $O(g_1^2 g_2 z^{-1})$. We find that the fourth-order graphs of the general type of (c) and (d) in Fig. 1, in which H' or H'' occurs in succession, cancel, leaving only the alternating fourth-order graphs of type (e) to contribute. Straightforward computation yields for the shift in ground-state energy, to second order in g_1,

$$\Delta E_G = -2g_1^2\left[1 + g_2 + \frac{g_2}{1-z^{-1}} + \frac{6}{5}g_2^2\right]\ . \tag{27}$$

Self-consistency is now imposed by equating $-\partial\Delta E_G/\partial g_1$ to 2μ and substituting from Eq. (26a). The determination of g_2 is simplified by using the mean-field value of g_0 in Eq. (26b) to approximate g_2 within the parentheses in Eq. (27), and by working only to $O(z^{-2})$ inside the parentheses. Thus, we obtain

$$\begin{aligned} 1 &\simeq 2(z-1)g_2\left[1 + 2g_2 + \frac{g_2}{z} + \frac{6}{5}g_2^2\right] \\ &\simeq 2(z-1)g_2\left[1 + \frac{1}{z} + \frac{4}{5}\frac{1}{z^2}\right] \\ &\simeq 2g_2\left[z - \frac{1}{5z}\right]\ . \end{aligned} \tag{28}$$

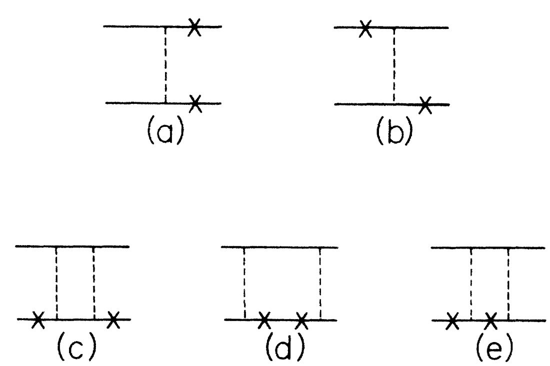

FIG. 1. Representative ground-state graphs, in the Bethe approximation, to third [(a) and (b)] and fourth [(c), (d), and (e)] order. The two solid lines represent a pair of neighboring grains and the dashed lines correspond to their Josephson coupling. The crosses indicate the mean field due to the other $z-1$ neighboring grains.

Combined with Eq. (26b), this yields

$$g_0^c = a_2 z^{-2} + a_3 z^{-3} + a_4 z^{-4} \quad (29)$$

of the desired form $\sum_{n=2}^{\infty} a_n z^{-n}$. As explained above, a_2 and a_3 retain their mean-field values of $\frac{1}{2}$ and 0, respectively. The result of our fourth-order computation, namely, $a_4 = \frac{3}{5}$, predicts, according to Eqs. (18) and (19), a fractional decrease in $R_\square^c$ of $0.6z^{-2}$ or, for $z = 4$, approximately 4%. Going beyond the Bethe approximation, we find[16] that for an open chain of N grains interacting among themselves according to H'', and interacting with the mean field of their neighbors according to H',

$$a_4(N) = \frac{6}{5} \frac{N-1}{N} . \quad (30)$$

For a closed chain of N grains, we find[16] the N-independent value $a_4 = \frac{6}{5}$. These various approximations to the complete interacting many-body system indicate that the error in Eq. (21) due to mean-field theory may amount to 8% or even 10%, but probably not significantly more than this. It would obviously be useful to compute a_4 for large clusters having a shape that is more two dimensional.[17]

To summarize, the theory presented above predicts not only that there should be a universal sheet resistance but also yields a numerical value for $R_\square^c$ that is in satisfactory accord with the observed value. Of the three approximations involved, that of the mean field seems the least serious and most under control. This is because of the error estimate made possible by the expansion in inverse powers of the number of grain neighbors. The error entailed in neglecting the frequency dependence of ΔC is more uncertain but ought to be amenable to some theoretical study in the future. The most serious problem in refining the theoretical prediction for $R_\square^c$ will be taking the disorder of the granular film into account.

In closing, we are happy to acknowledge support by the National Science Foundation via Grant for Basic Research No. DMR 85-06009.

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[17] This would include the more conventional Bethe approximation, which would surround a central grain by non-mean-field neighbors.