First-Order Reentrant Transition in Granular Superconducting Films

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A quantum Monte Carlo method is developed to treat path integrals at finite temperature and with topological constraints to study a periodic model of a granular superconductor. The results obtained provide strong evidence for a low-temperature first-order reentrant transition induced by zero-point quantum fluctuations.

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The possibility of having a low-temperature reentrant phase transition in granular systems, induced by quantum fluctuations, has been a source of controversy for some time.[1-6] The first indications that a reentrant transition could appear in such systems came from mean field theory (MFT) and self-consistent harmonic approximations.[1] Later, further analysis of the MFT led to the conclusion that there is no reentrance in a model that includes only diagonal charging-energy effects and that there is a very restrictive kind of transition in the model that includes the off-diagonal terms.[2,3] Since there are fairly convincing arguments which show that there must be a nonzero critical value of the magnitude of quantum fluctuations below which the system is superconducting at zero temperature,[5] one may be led to surmise that no such transition should take place in these systems. Recently, however, one of us[6] has carried out a semiclassical calculation in the diagonal model in two dimensions. From this renormalization-group (RG) analysis, evidence for a low-temperature instability was found which could be identified with a reentrant transition. Although this analysis included fluctuations that were left out in the MFT treatments, the calculation is based on a perturbative expansion in Plank's constant. One would expect that these results should be valid close to the Kosterlitz-Thouless (KT) critical temperature, where long-range phase coherence sets in, but less so at lower temperatures, where a reentrant transition may take place. To resolve this controversy, it is therefore necessary to study this problem nonperturbatively. The purpose of this paper is to present the results of the first such calculation.

Our method of attack consisted in developing an appropriate Monte Carlo (MC) algorithm designed specifically for the diagonal model. From the results of this analysis we conclude that *there is indeed* a low-temperature instability in the model which can be interpreted as a reentrant transition. Further, our MC data provide strong evidence showing that this transition is, in fact, first order.

The model we study is defined in terms of the imaginary-time partition function[6]

$$Z=\int d[\phi(\tau)]\exp\left\{-\frac{1}{\hbar}\int_0^{\hbar\beta}\sum_i\left[\frac{\hbar^2}{2u}\left(\frac{d\phi_i(\tau)}{d\tau}\right)^2+E_J\Delta_i\phi(\tau)\right]d\tau\right\}, \tag{1}$$

where $\Delta_i\phi(\tau)=1-\cos[\phi_{i+1}(\tau)-\phi_i(\tau)]$. In (1), $\phi\in[0,2\pi]$ is the phase of the order parameter, u measures the strength of quantum effects, and E_J is the Josephson tunneling amplitude, which we take to be independent of temperature; $\beta=1/k_BT$ with T the temperature, and the sum over i is over two-dimensional vectors defined on a square lattice. There are several important differences between the above partition

function and those studied in usual MC simulations. First, notice that the temperature appears in the limits of integration over imaginary time, and also in the essential quantum-periodicity condition, $\phi(\tau+\beta\hbar)=\phi(\tau)$. Because ϕ takes values in the circle this imposes a topological constraint in the path integral which has to be kept track of. To perform the MC simulation, we discretize the imaginary-time direction; then the correct low-temperature quantum results arise when the continuum limit in this direction is taken. As we discuss below, this necessitates a rather large amount of computing.

To illustrate this procedure, consider first the case $E_J=0$. This corresponds to the trivial $d=0$ quantum-mechanical problem of a set of decoupled rigid rotors, for which an exact answer is known. We want to rederive this answer from the path integral given above. Discretizing (1) with $E_J=0$ we get for each rotor

$$Z_0=(L_\tau/2\pi\beta u)^{L_\tau/2}\int d[\phi]\exp[-\sum_\tau(L_\tau/2\beta u)\Delta_\tau\phi(\tau)], \tag{2}$$

where $\Delta_\tau\phi(\tau)=1-\cos[\phi(\tau+1)-\phi(\tau)]$. The periodicity of ϕ is taken care of by the form that we have chosen for $\Delta_\tau\phi$. In (2) we have divided the time interval into L_τ time slices with separation ϵ such that $L_\tau\epsilon=\beta\hbar$. The continuum is recovered by considering the limit $L_\tau\to\infty$, $\epsilon\to 0$, with $L_\tau\epsilon$ fixed. Notice that the action in (2) looks like that of a classical XY model in one dimension. The difference here, however, is that at finite temperatures the evaluation of (2) requires that the quantum-periodicity constraint on ϕ be taken into account. Therefore, the known solutions of the $d=1$ XY model do not correspond to the quantum-rotor solutions in the continuum limit. An analytic evaluation of Z_0 leads to the correct rotor solutions in the continuum limit. Our interest here is to obtain such solutions using MC methods.

As in MC calculations in the classical XY model, we approximate the U(1) symmetry of the problem by considering a Z(N) subgroup. However, here we found that in order to obtain reliable results for the quantum rotor, we needed N much larger than usual: Whereas $N\approx 12$ is sufficient in the classical case (at moderate temperatures), here we needed $N\approx 5000$. More important is the fact that L_τ has to be large at low temperatures. For instance, for $\beta u=30$ we needed $L_\tau=1500$ in order to get results differing from the exact answers by 2%. Also as important is the fact that we needed a rather large number of MC steps (MCS) per site to reach thermodynamic equilibrium. Of course, this $d=0$ problem requires larger values of L_τ than a higher-dimensional problem. Nevertheless, the $d=0$ problem does provide lessons about the temperature dependence of the number of levels, L_τ, necessary to obtain continuum-limit answers, as well as the number of MCS/site required.

We can now turn to the problem defined by Eq. (1), which is the central topic of this paper. The discrete version of (1) reads

$$Z=\left(\frac{L_\tau}{2\pi\alpha K}\right)^{V/2}\int d[\phi]\exp\left\{-\sum_{x,y,\tau}\left[\frac{L_\tau}{K\alpha}\Delta_\tau\phi_i(\tau)+\frac{K}{L_\tau}\Delta_i\phi_i(\tau)\right]\right\}, \tag{3}$$

with $\Delta_i\phi$ and $\Delta_\tau\phi$ defined below (1) and (2). In (3) we have defined $V=L_xL_yL_\tau$, with L_x,L_y the spatial lattice dimensions, $K=\beta E_J$, and we have defined the parameter, $\alpha=u/E_J$, which measures the relative importance of quantum to thermal fluctuations in the model. To see continuum results we must have $L_\tau>>K\alpha=\beta u$. In this representation we recognize an important dual symmetry with a clear physical content: At low temperatures the kinetic (or charging-energy) term dominates over the potential (or Josephson) contribution, with the opposite true at high temperatures. This dual symmetry is crucial in the construction of an efficient MC algorithm since one term is relevant when the other is not. We are mainly interested in the ultralow-temperature regime, so we carry out the MC updating in two steps: First we go through our three-dimensional lattice shifting an entire column of ϕ's along the τ direction by the ame angle, accepting the update in the usual MC manner. Next we go through each site updating individual angles using a standard Metropolis algorithm. This method, entailing two separate updates, was found to be very efficient at low temperatures.

We calculated several thermodynamic quantities as functions of K, α, the volume in our slab geometry, and the number of MCS/site. Here we shall report on the results obtained for the helicity modulus, Υ, the internal energy, E, and the specific heat, C. From the response of the system to a twist along one of the spatial directions we derive the ap-

propriate expression for Υ following the logic of Ohta and Jasnow.[7] This quantity provides a good test of the superfluid properties of the system since it is directly proportional to the superfluid density.[7] We first checked our calculations in the classical regime and indeed found that Υ has a rapid variation around the KT critical temperature, defined as the temperature at which Υ intersects the line passing through the origin with slope $2/\pi$. From RG analysis[6] we know that universality of the critical exponent η still holds for small enough α. How small α should be, though, is not known. In Fig. 1 we show our MC results for the KT critical temperature as a function of α. Each set of values for Υ (10–12 for each value of α) was obtained by averaging over 200 sets of 200 iterations for a total of 4×10^4 MCS/site (after discarding 10^4 MCS/site for thermalization). A linear least-squares fit to the data up to $\alpha=2$ gives $1/K_c=0.930\,36-0.088\,15\alpha$, which agrees rather well with the $\alpha=0$ (classical) value. The smallness of the coefficient of the linear term supports our earlier assertion about the correctness of the semiclassical approximation of quantum effects close to the KT critical region. As α increases, the specific heat has a hump of decreasing height and at a decreasing temperature, which is, as in the classical case, always above $1/K_c(\alpha)$.

We now come to the central results of this paper, which pertain to the ultralow-temperature region of the model. In Fig. 2 we show results for the helicity modulus (normalized by dividing by $a^2L_xL_yE_J$, with a the physical lattice spacing) and the specific heat (normalized by dividing by $k_BL_xL_yE_J^2$) for $\alpha=0.3$ and $E_J/\hbar=1$. Each point was measured by averaging over 200 sets of 200 iterations each, for a total of 4×10^4 MCS/site (after discarding 10^4 MCS/site). Some consistency checks were performed with longer runs (as long as 3×10^5 MCS/site). Starting at around $1/K=0.1$, both Υ as well as C increase smoothly as the temperature is decreased. This smooth behavior continues up to $1/K\approx0.03$, at which point there is a sharp drop in both of these quantities as a function of temperature. This behavior is markedly different from that observed in the classical case,[7] and is therefore fundamentally due to quantum-mechanical effects. This dramatic change in behavior for both Υ and C is a clear signal for the existence of a reentrant transition. To determine the nature of this transition we looked at the values of several quantities as functions of the MC iteration number. A particularly revealing example of this analysis is provided by the internal energy. In Fig. 3 we show the results for the MC time evolution of the internal energy in the region $0.01\leq1/K<0.03$ for $\alpha=0.3$. (Each point represents 200 iterations, for a maximum of 4×10^4 MCS/site). These results provide strong evidence for the existence of two different stable phases in this region and, hence, for the existence of a first-order transition; a totally unexpected result. Further evidence supporting this conclusion comes from the observed metastability in this region: We were unable to obtain reliable results for either the specific heat or the helicity modulus (both quantities being measured by averaging fluctuations). It is possible that a careful pruning of the data may lead to results for these quantities in the critical region, but no such effort was made. We have not as yet performed a systematic analysis of finite-size effects, and these effects may be crucial in determining conclusively the

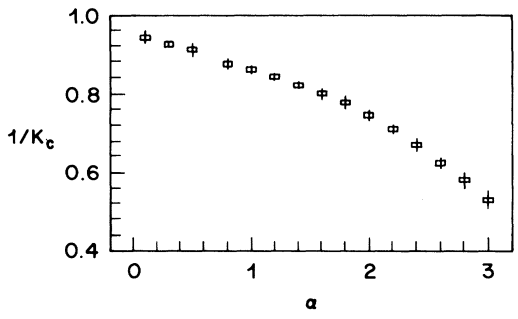

FIG. 1. KT critical temperature as a function of α. Lattice sizes were $10\times10\times L_\tau$, with $L_\tau=10$ for $0\leq\alpha\leq2$ and $L_\tau=30$ for $\alpha>2$.

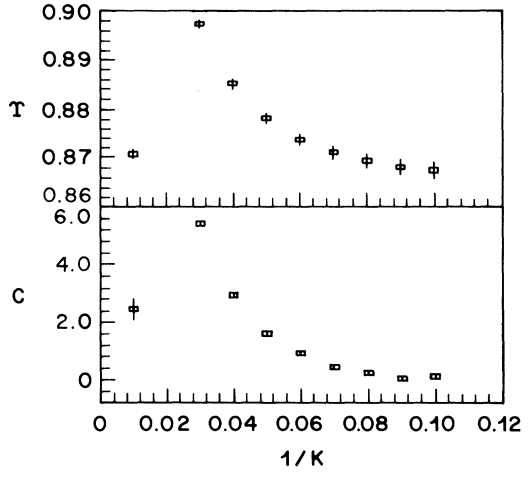

FIG. 2. Helicity modulus, Υ, and specific heat, C as functions of $1/K$ for $\alpha=0.3$. Lattice sizes were $16\times16\times L_\tau$, with $L_\tau=40$ for $1/K\geq0.06$, $L_\tau=60$ for $0.03\leq1/K<0.05$, and $10\times10\times150$ for $1/K\leq0.03$.

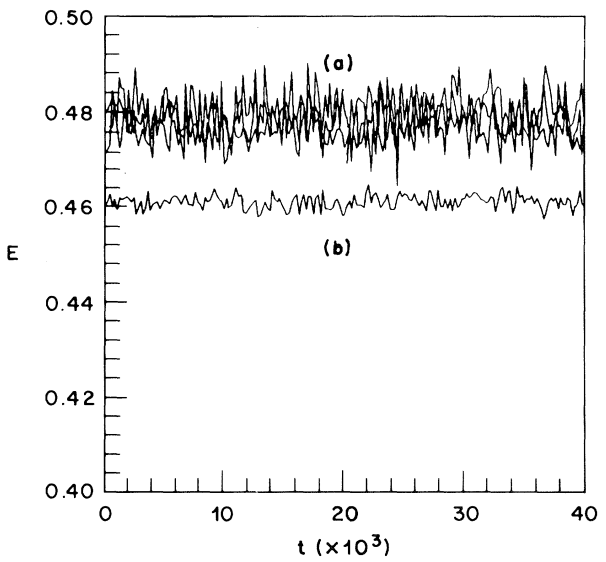

FIG. 3. Internal energy per site (divided by E_J) as a function of the MC iteration number for $\alpha = 0.3$. The three upper sets of points (labeled a) correspond to $1/K = 0.015$, 0.020, and 0.025; the lower set of points (b) corresponds to $1/K = 0.010$. All lattices sizes were $10 \times 10 \times 150$.

order of the transition that we have found.

We can argue about the nature of the phenomenon which triggers the low-temperature transition. From RG analysis[6] it follows that quantum fluctuations nucleate vortex pairs as the temperature decreases. The fact that thermal vortex pairs also nucleate as the temperature decreases implies that the fugacity should have a minimum as a function of temperature, leading to a first-order transition.

To conclude, we have presented, for the first time, strong nonperturbative evidence pointing to the existence of a reentrant transition in a periodic model of a granular system. The transition is induced by quantum fluctuations and is found to be first order. Experimentally, reentrance may be seen in regular arrays of Josephson junctions in which the charging energy is large and can be varied,[8] or in granular systems in which the grains are small and the samples are not too inhomogeneous.[9-11]

Most of the numerical work involved in this analysis was done on a CRAY-1 computer. A total of approximately 40 h of CPU time was used. More details about the calculations as well as more extensive results will appear elsewhere.[10]

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[1]E. Simanek, Phys. Rev. B **23**, 5762 (1981).

[2]K. B. Efetov, Zh. Eksp. Teor. Fiz. **78**, 2017 (1980) [Sov. Phys. JETP **51**, 1015 (1980)].

[3]P. Fazekas, Z. Phys. B **45**, 215 (1982); Y. Imry and M. Strongin, Phys. Rev. B **24**, 6353 (1981); M. Schroter, P. Fazekas, and B. Muhlschlegel, to be published.

[4]D. M. Wood and D. Stroud, Phys. Rev. B **25**, 1600 (1982).

[5]S. Doniach, Phys. Rev. B **24**, 5063 (1981).

[6]J. V. José, Phys. Rev. B **29**, 2836 (1984).

[7]T. Ohta and D. Jasnow, Phys. Rev. B **20**, 139 (1979).

[8]R. Voss and R. Webb, Phys. Rev. B **25**, 3446 (1983).

[9]S. A. Wolf, D. U. Gubser, W. W. Fuller, G. C. Garland, and R. S. Newrock, Phys. Rev. Lett. **47**, 1071 (1981).

[10]L. Jacobs, J. V. José, and M. A. Novotny, to be published.

[11]After this Letter was submitted for publication, we learned that preliminary supporting experimental evidence has been found in ultrathin tin films by A. Goldman *et al.* (A. Goldman, private communication).