The superconducting phase diagram in a model for tetragonal and cubic systems with strong antiferromagnetic correlations

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We calculate the superconducting phase diagram as a function of temperature and z-axis anisotropy in a model for tetragonal and cubic systems having strong antiferromagnetic fluctuations. The formal basis for our calculations is the fluctuation exchange approximation (FLEX) applied to the single-band Hubbard model near half-filling. For nearly cubic lattices, two superconducting phase transitions are observed as a function of temperature with the low-temperature state having the time-reversal symmetry-breaking form, $d_{x^2-y^2} \pm id_{3z^2-r^2}$. With increasing tetragonal distortion the time-reversal-symmetry-breaking phase is suppressed giving way to only $d_{x^2-y^2}$ or $d_{3z^2-r^2}$ single-component phases. Based on these results, we propose that CeIn₃ is a candidate for exhibiting a time-reversal symmetry-breaking superconducting state.

The discovery of unusual superconductivity in the heavy fermion systems focused much early effort on electronic pairing mechanisms and the possibility that the resulting superconducting states have lower symmetry than the underlying crystalline lattice. The competition between superconductivity and magnetic ordered states suggests that spin fluctuations are a prime candidate to form the glue that binds electrons into Cooper pairs. Power law dependencies of thermodynamic properties and phase diagrams that include multiple superconducting states, for example the $H-T$ phase diagram of UPt₃ and the $x-T$ phase diagram of U₁₋ₓThₓBe₁₁₃, point to the lower symmetry of the superconducting order parameter. As a consequence of having a lower symmetry in comparison to that for the lattice, the order parameter of an unconventional superconductor can have more degrees of freedom than exhibited in a conventional superconductor leading to phase diagrams involving multiple superconducting states, but microscopic models are needed to shed light on the connection between the pairing interaction and the pairing states that are produced.

Recently, the interplay between crystal lattice symmetry and spin-fluctuation induced pairing instabilities was explored by Monthoux and Lonzarich and Arita, Kuroki and Aoki. They find that spin fluctuation-induced pairing into the unconventional $d_{x^2-y^2}$ state is most effective for producing large transition temperatures ($T_c$) for quasi two-dimensional lattices, a result that is consistent with the observation that the highest $T_c$ values occur in the quasi two-dimensional cuprates and the more recent finding that $T_c$ in the cubic heavy fermion system CeIn₃ is about one order of magnitude less than is observed in a collection of related quasi two-dimensional compounds, such as the series Ce₉TₘInₙSn₂m where $T =$ Rh or Ir and $n = 1$ or $2$ and $m = 1$.  

In this Letter we report on model calculations of the superconducting phase diagram as a function of temperature and tetragonal anisotropy for electrons paired via spin fluctuations in nearly antiferromagnetic systems. The phase diagram is obtained numerically through the proper generalization of the fluctuation exchange approximation for the single band Hubbard model to the superconducting state, a generalization which is necessary to resolve the relative stability of nearly degenerate unconventional pairing states. We find that for cubic lattices the stable superconducting state has the time-reversal symmetry breaking form $d_{x^2-y^2} \pm id_{3z^2-r^2}$. Small tetragonal distortions lift the degeneracy between $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ pairing states leading to two superconducting phase transitions as a function of decreasing temperature, but as the degeneracy is lifted further through larger tetragonal distortions the second transition is suppressed. On the basis of these results, we suggest that the low-temperature superconducting state in simple cubic systems with strong antiferromagnetic correlations, such as CeIn₃, are strong candidates for realizing multiple superconducting phases including a low-temperature phase with broken time-reversal symmetry.

The microscopic basis for our calculations is the single-band Hubbard model,

$$
H = -\sum_{i,j,\sigma} \left( t_{ij} c_{i,\sigma}^\dagger c_{j,\sigma} + \text{h.c.} \right) + U \sum_i c_{i,\uparrow}^\dagger c_{i,\uparrow} c_{i,\downarrow}^\dagger c_{i,\downarrow},
$$

where $t_{ij}$ represents the electron hopping amplitude between sites $i$ and $j$ and $U$ is the on-site interaction energy between up and down spin electrons. The values of $t_{ij}$ reflect the underlying lattice structure. For tetragonal lattices, the hopping parameter for unit displacements in the $x$ or $y$ directions, $t_{xy}$, is distinct from the same for unit displacements along the $z$ axis, $t_z$. For simplicity we set the hopping integrals equal to zero for larger displacements in which case the non-interacting electron bandwidth is equal to $W_0 = 8t_{xy} + 4t_z$.

An approximation scheme must be employed for calculations based on this model. We use the fluctuation...
exchange approximation (FLEX) of Bickers, White, and Scalapino, a numerically-based scheme that is conserving in the sense described by Baym. FLEX provides a self-consistent description of both the quasiparticles and the magnetic-fluctuation induced pairing interaction for a given on-site interaction strength, temperature, and band filling. It is expected that results obtained with FLEX are quantitatively accurate for, at best, weak-to-intermediate coupling, i.e. \( U/W_0 \lesssim 1 \). However, even within this range there are notable qualitative failures such as, as will be discussed shortly, violations of the Mermin-Wagner-Hohenberg theorem for \( d \)-wave superconducting states.

In contrast to other works, our formulation of FLEX includes the entire set of fluctuation diagrams for the electron self-energy in both the normal and superconducting states, i.e. we include all possible combinations of particle-like, hole-like and anomalous Green’s functions. At the formal level, this ensures that response functions and Green’s functions are obtained in a consistent manner such that conservation laws derivable from symmetries of the Hamiltonian are obeyed. While \( T_c \) is determined by self-energy diagrams which contain only one anomalous Green’s function, a more complex set of diagrams with three anomalous Green’s functions contribute to fourth order terms in the Ginzburg-Landau expansion for the free energy in terms of the superconducting order parameter and these terms are known to determine the relative stability of multicomponent pairing states. We note that the results we obtain for \( T_c \) in two dimensions are in agreement at the 10% level with those obtained earlier suggesting that self-energy diagrams that are omitted in most treatments of FLEX do not play a large role in determining \( T_c \), at least in the two-dimensional limit.

To access sufficiently large system sizes with modest computational resources, we combine this formulation of FLEX with the dynamical cluster approximation (DCA), essentially, in the DCA large lattice calculations are made feasible by approximating correlation effects via a smaller embedded cluster. When combined with the DCA, there are three numerical parameters in FLEX: the number of Matsubara frequency points used \( m \), the lattice size \( N_L \) and the DCA cluster size \( N_c \). We show the dependence of results for the pairing amplitude, \( m_p \) (defined below), versus temperature on these numerical parameters in Figure 1 for a two-dimensional lattice. For the range of these parameters that we can access, the most significant variation in these curves is through the DCA cluster size (bottom graph in Figure 1). Nonetheless, we find that the error made in the result for \( T_c \) is only on the order 10% when using a \( 4^2 \) DCA cluster. In what follows, we use \( m = 16384 \) frequency points, \( N_L = 32^3 \) lattices and \( N_c = 4^3 \) DCA clusters.

Although a variety of electronic and thermodynamic properties can be calculated with the FLEX, our main focus here is on results for the superconducting transition temperature and the associated order parameter symmetry. We obtain these results by determining the spatial dependence of the pair wave function, \( \psi(\mathbf{r}) \), for the stable superconducting state and the associated pairing amplitude, \( m_p \). These quantities are obtained from the self-consistent result for the anomalous Green’s function, \( F \), via

\[
F_{\uparrow\downarrow}(\mathbf{r} \rightarrow 0^-, \mathbf{r}) \equiv \langle c_{\mathbf{r}=0,\uparrow} c_{\mathbf{r},\downarrow} \rangle = m_p \psi(\mathbf{r}).
\]

Other than requiring that \( \psi(\mathbf{r}) \) is even as a function of \( \mathbf{r} \) (i.e. we restrict ourselves to singlet-pairing), no assumption is made on the symmetry of the pairing state. To permit any possible singlet-pairing state to emerge in these calculations, we initialize the self-consistent FLEX equations with a small, spatially random pairing field to induce a pairing amplitude in all possible symmetry channels. The small field is removed as the self-consistent procedure projects the wavefunction of the most stable pairing state, \( \psi(\mathbf{r}) \).

We emphasize the mean-field nature of the FLEX phase diagram for \( d \)-wave superconductivity in the single-band Hubbard model. The mean-field \( T_c \) values essentially represent the temperature at which superconducting order emerges locally. The true thermodynamic \( T_c \) is determined by phase fluctuations that are not present in FLEX. The neglect of these fluctuations is especially problematic in the two-dimensional limit where they are known to eliminate the possibility of a finite temperature phase transition. Nonetheless, it is well known that a relatively weak interplanar coupling can stabilize superconductivity in a quasi-two-dimensional systems and the mean-field phase diagram is revealing with respect to determining the conditions under which the tendency toward superconducting order is greatest.

Our primary result, shown in Figure 2 is the calculated superconducting phase diagram as a function of the scaled temperature, \( k_B T/W_0 \), and the ratio of interplanar to intraplanar hopping, \( t_z/t_{xy} \) for fixed values of density \( n = 0.85 \) electrons per site and the ratio of the on-site interaction energy to bare electron bandwidth \( (U/W_0 = 0.5) \). We focus on three features in Figure 2: the low-temperature stability of the time-reversal symmetry breaking state \( d_{xz,yz}^{+} \pm i d_{z2-\tau^z} \) for lattices with cubic and nearly cubic symmetry, two distinct superconducting transitions as a function of temperature for nearly cubic lattices, and, in agreement with previous results, maximal \( T_c \) values occurring in the quasi-two-dimensional limit.

We first focus on the cubic limit, \( t_z/t_{xy} = 1 \), for which \( T_c \) is at a local minimum. We find that, in agreement with Arita, Kuroki and Aoki, that the stable superconducting state belongs to the two-fold degenerate representation \( \Gamma_3^\pm \) of the cubic group which is described by basis functions of the form \( d_{xz,yz}^{\pm} \) and \( d_{z2-r_z} \). Symmetry considerations allow for either single-component
or multicomponent states of the form $d_{x^2-y^2}$, $d_{3z^2-r^2}$, $d_{x^2-y^2} \pm d_{3z^2-r^2}$, or $d_{x^2-y^2} \pm id_{3z^2-r^2}$. Weak-coupling-based arguments suggest that the most stable state is the one for which the superconducting gap is most complete on the Fermi surface as this will tend to maximize the condensation energy. This argument favors the $d_{x^2-y^2} \pm id_{3z^2-r^2}$ pairing state as it has point nodes while the others have line nodes \[12\].

FLEX incorporates feedback effects between quasiparticles, the order parameter and the pairing interaction and, thus, does not necessarily generate the pairing state expected from weak-coupling theory. Nonetheless, we indeed find that FLEX produces the $d_{x^2-y^2} \pm id_{3z^2-r^2}$ pairing state in this case. This state breaks time-reversal symmetry leading to unusual phenomena such as bulk magnetic effects associated with the superconducting pairs \[1\]. Assuming that quasiparticles are well-defined in the superconducting state, such pairing is expected to generate thermodynamic properties that reflect the point node structure of the gap function, such as having a $T^3$ low-temperature specific heat.

For slight deviations from cubic symmetry, i.e. $t_z/t_{xy} = 1 \pm \epsilon, \epsilon \ll 1$, $T_c$ increases due to the improved stability of either the $d_{x^2-y^2}$ or $d_{3z^2-r^2}$ pairing state. The two pairing states are no longer degenerate for $\epsilon \neq 0$ and the initial transition from the superconducting transition is into a single component state. However, because of the near degeneracy of these states, a second superconducting transition occurs into the $d_{x^2-y^2} + id_{3z^2-r^2}$ state with decreasing temperature. The low-temperature state is nodeless because $d_{3z^2-r^2}$ basis functions are members of the symmetric group for tetragonal lattices and, consequently, are intrinsically mixed with contributions from basis functions with s-wave symmetry. However, s-wave terms in the pair wavefunction are relatively small. For example, the s-like terms have amplitudes which are about 4% of those for $d_{3z^2-r^2}$ for $t_z/t_{xy} = 1.1$. Consequently, the gap is correspondingly small for a set of points on the Fermi surface that correspond to the point nodes that are found for cubic lattices.

As is expected, we find that for lattices favoring in-plane conduction, i.e. $0 \leq t_z/t_{xy} < 1$, the $d_{x^2-y^2}$ pairing state is most stable, while the $d_{3z^2-r^2}$ ($+$ s-wave) pairing state is most stable when interplanar motion is enhanced, i.e. $t_z/t_{xy} > 1$. It is apparent from Figure \[2\] that the largest $T_c$ values are obtained in the quasi-two-dimensional limit, i.e. $t_z/t_{xy} \rightarrow 0$, in agreement with the results referred to earlier \[2, 8\] and in line with the trend observed in the compounds CeIn$_3$ \[8\] and Ce$_n$T$_m$In$_{3n+2m}$ where $T = \text{Rh or Ir and}$ $n = 1$ or 2 and $m = 1 \ 8$. There also is a region of enhanced $T_c$ for $t_z/t_{xy} > 1$ up to approximately $t_z/t_{xy} \sim 2$ at which point $T_c$ becomes vanishingly small. We note that $t_z/t_{xy} = 2$ corresponds to the point at which the interplanar bandwidth equals the bandwidth corresponding to in-plane motion.

It is interesting to consider these results in light of experimental data for superconductors with cubic symmetry. Data for the compound PrO$_4$Sb$_{12}$ is consistent with the superconducting phases observed in this calculation \[8\]. For example, specific heat measurements show evidence of a double superconducting transition \[14\] and muon-spin relaxation data is consistent with a low-temperature time-reversal symmetry breaking state \[16\]. However, the strong collective mode that forms the glue between paired electrons is due, most likely, to electric quadrupolar rather than magnetic degrees of freedom \[16\]. Nonetheless, it is interesting to note the possibility that two different pairing mechanisms may tend to generate the same time-reversal symmetry breaking state in cubic symmetry and, thus, the phenomenology developed for the superconducting state in PrO$_4$Sb$_{12}$ \[16\] may apply also for magnetically-paired superconductors.

The alloy series U$_{1-x}$Th$_x$Be$_{13}$ displays a complex superconducting phase diagram as a function of thorium concentration, $x$. For $0.2 \lesssim x \lesssim 0.4$, a double superconducting transition is observed in the electron specific heat \[17\] and muon spin resonance data points to the appearance of an internal magnetic field below the second transition. Sigrist and Rice interpreted the magnetic anomaly in terms of a multicomponent time-reversal symmetry breaking superconducting state \[15\]. However, Kromer et al. \[16\] use specific heat and lattice expansion data to argue for the appearance of spin density wave below $T_c$. The interpretation of the phase diagram for these compounds is still being debated \[20\].

We simply note that the scenario described by Sigrist and Rice is consistent with pairing states that are observed in the model that we consider here and should their interpretation be incorrect it would point to the importance of processes that are neglected in spin-fluctuation models.

The cubic superconductor CeIn$_3$ is, perhaps, the most likely candidate system for connecting with these model calculations. On account of the vicinity of the Neél state, there is a strong likelihood that electron pairing is related to the strong antiferromagnetic correlations in this system. To the best of our knowledge, no evidence has been presented either for a double superconducting transition as a function of temperature nor for a low-temperature time reversal symmetry breaking state in this compound. In light of weak coupling arguments \[12\] the FLEX results presented here and the anomalies observed in other unconventional cubic superconductors, the presence or absence of time-reversal symmetry breaking in this compound will be revealing with respect to the applicability of simple models, such as the one considered here, for providing a minimal microscopic basis for understanding superconductivity in CeIn$_3$ and related compounds.

In summary, we have performed fluctuation exchange approximation calculations for the Hubbard model near half-filling to model the phase diagram of tetragonal and cubic superconducting systems whose pairing is mediated by antiferromagnetic spin fluctuations. Near cubic sym-
pairing states are most stable for $t/U/W = 0.85$ electrons/site and with broken time reversal symmetry. On the basis of the second transition occurs to a multicomponent state, which corresponds to a transition to a single-component state and symmetry breaking state. For $t > t_{xy}$, a second transition occurs as a function of decreasing temperature into the time-reversal symmetry breaking state $d_{x^2-y^2} + i d_{3z^2-r^2}$.

In these calculations, we propose that CeIn$_3$ is a candidate material for exhibiting broken time-reversal symmetry in the superconducting state.

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[1] M. Sigrist and K. Ueda, Rev. Mod. Phys. 63, 239 (1991).
[2] P. Monthoux and G. G. Lonzarich, Phys. Rev. B 59, 14508 (1999).
[3] R. Arita, K. Kuroki, and H. Aoki, Phys. Rev. B 60, 14585 (1999).
[4] F. Grosche, S. R. Julian, N. D. Mathur, and G. G. Lonzarich, Physica B 223-224, 50 (1996).
[5] J. D. Thompson, Movshovich, Z. Fisk, F. Bouquet, N. J. Curro, R. A. Fishere, P. C. Hammel, H. Hegger, M. F. Hundley, M. Jaime, et al., Journal of Magnetism and Magnetic Materials 226-230, 5 (2001).
[6] N. E. Bickers, D. J. Scalapino, and S. R. White, Phys. Rev. Lett. 62, 961 (1989).
[7] G. Baym, Phys. Rev. 127, 1391 (1962).
[8] N. E. Bickers and S. R. White, Phys. Rev. B 43, 8044 (1991).
[9] J. K. Freericks, Phys. Rev. B 50, 403 (1994).
[10] C.-H. Pao and N. E. Bickers, Phys. Rev. B 49, 1586 (1994).
[11] M. H. Hettler, M. Mukherjee, M. Jarrell, and H. R. Krishnamurthy, Phys. Rev. B 61, 12739 (2000).
[12] M. Sigrist, Physica C: Superconductivity 341-348, 695 (2000).
[13] J. Goryo, Phys. Rev. B 67, 184511 (2003).
[14] R. Vollmer, A. Faißt, C. Pfleiderer, H. v. Löhneysen, E. D. Bauer, P.-C. Ho, V. Zapf, and M. B. Maple, Phys. Rev. Lett. 90, 057001 (2003).
[15] Y. Aoki, A. Tsuchiya, T. Kanayama, S. R. Saha, H. Sugawara, H. Sato, W. Higemoto, H. Koda, K. Ohishi, K. Nishiyama, et al., Phys. Rev. Lett. 94, 067003 (2003).
[16] M. Kohgi, K. Iwasa, M. Nakajima, N. Metoki, S. Araki, N. Bernhoeft, J. Mignot, A. Gukasov, H. Sato, Y. Aoki, et al., J. Phys. Soc. Jap. 72, 1002 (2003).
[17] H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. B 31, 1651 (1985).
[18] M. Sigrist and T. M. Rice, Phys. Rev. B 39, 2200 (1989).
[19] F. Kromer, D. Helfrich, M. Lang, F. Steglich, C. Langhammer, A. Bach, T. Michels, J. S. Kim, and G. R. Stewart, Phys. Rev. Lett. 81, 4476 (1998).
[20] V. Martisovits, G. Zarand, and D. L. Cox, Phys. Rev. Lett. 84, 5872 (2000).