Effect of matrix elements on the pairing kernel in heavy-fermion superconductors

M. R. Norman
Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439
(Received 17 February 1993; revised manuscript received 17 May 1993)

A method is derived for calculating the pairing kernel in heavy-fermion superconductors including matrix-element effects. Various models for the interaction vertex are considered, including pseudospin exchange, orbital exchange, and quadrupolar exchange. As an example, this formalism is applied to UPt_3 with the use of relativistic wave functions from a local-density-band calculation.

Much work has been done over the past decade in trying to understand the microscopic basis for superconductivity in heavy-fermion metals, and more recently in the copper oxides. In the heavy-fermion case, a large body of experimental data points to the presence of a nontrivial order parameter reminiscent of ^{3}He. Because of this, many theorists working on this subject, including the author, have borrowed techniques successful for ^{3}He and applied them to the heavy-fermion problem. In particular, a favorite mechanism being explored is pairing due to spin fluctuations. In the calculations performed, one uses some model for the dynamical susceptibility and solves a gap equation. The solution is determined by the k dependence of the susceptibility and the Fermi surface.[1-4] Similar calculations have been recently performed on copper oxide materials.[5]

The deficiency of this approach is obvious since it assumes that the pair vertex is a simple function of momentum transfer, $V(k-k')$, usually taken to be proportional to the experimental susceptibility. For multiband systems like heavy fermions, though, one requires the decomposition $V_{nn'}^{\alpha\beta;\gamma\delta}(k-k')$ to properly solve the gap equation, where n and n' are band indices and α, β, γ, and δ are pseudo-spin indices. This point was realized from the beginning of theoretical work on this subject.[6] Despite this, little work has been done along these lines, a noticeable exception being a paper by Appel and Hertel,[7] where a real space approach to pairing in heavy fermions is advocated. An advantage to their approach is that the proper dependence on band and pseudospin indices is insured by taking the pair vertex in real space and Fourier transforming to momentum space using wave functions from a band-structure calculation. Since it is the momentum dependence which determines the symmetry of the gap function, and since renormalizations in heavy fermions are primarily frequency dependent in character, the band wave functions should be sufficient for determining relative coupling constants.

In this paper, following the spirit of Refs. 6 and 7, the author develops a formalism for calculating the pairing kernel in heavy-fermion superconductors including relativistic matrix elements determined from band-structure calculations. Various models for the exchange part of the kernel are considered, including pseudospin, orbital, and quadrupolar exchange. As an example, this method is applied to the case of UPt_3, the best studied of the heavy-fermion superconductors.

The pairing kernel to be evaluated is of the form

$$\langle k\alpha,-k\beta|V|k'\gamma,-k'\delta\rangle ,$$

where V is the pairing interaction (note that the band index is implicitly included in the definition of k). This form reduces to those considered in Refs. 1–5 if $|k\alpha\rangle$ is taken to be a free-electron plane wave times a spinor and V is a function of $r-r'$. In this paper, a different approach is taken by using the results of local density band calculations for $|k\alpha\rangle$. Moreover, since it is assumed that the physics is primarily determined by the f electrons, an approximation is made of assuming that V acts only on the $J=\frac{5}{2}f$ states on the f atom site. V is further approximated by assuming it has just three values corresponding to whether the two electrons are on the same site (U), on near-neighbor sites (J), and on next-near-neighbor sites (JN). [A more sophisticated approach would be based on a microscopic model for $\chi(r,r')$.] Finally, the dependence on orbital quantum number, μ (where μ ranges from $-\frac{5}{2}$ to $\frac{5}{2}$), is taken into account by the exchange operator, denoted by X, with V taking the form $V_{RR'}^{ij}X_iX_j$ where i and j represent Cartesian indices (x,y,z), R and R' denote the positions of f atom sites, and X will be some appropriate analogue of the Pauli spin matrix, σ. Given this, the pairing kernel is now of the form

$$\sum_{R,R',i,j} V_{RR'}^{ij}\langle k\alpha|X_i|k'\beta\rangle_R\langle -k\gamma|X_j|-k'\delta\rangle_{R'} , \tag{1}$$

where $\langle|\rangle_R$ denotes an integral over a Wigner-Seitz sphere centered at R. To evaluate this, note that the band-structure wave functions are of the form

$$|k\alpha\rangle=\sum_{\mu,R} a_{\mu,k,R}^{\alpha}|\mu\rangle_R , \tag{2}$$

where $|\mu\rangle$ is the appropriate combination of spherical harmonics and spinors times the f electron radial function. Since X has no dependence on the radial coordinate, the radial integrals factor out of the problem [except that since a linear muffin-tin-orbital (LMTO) band method is used, the wave function has a radial term and its energy derivative as expansion functions. For notational convenience, this complication is ignored, but is taken into account when evaluating the matrix elements]. In a band calculation, $|k\alpha\rangle$ is just generated in one irreducible wedge of the zone and only on the sites inside the primitive cell. The rest of the information for $|k\alpha\rangle$ can

 ©1993 The American Physical Society

be obtained by use of Bloch's theorem plus by application of the group operations on $|k\alpha\rangle$. This is somewhat complicated for UPt_3 since the space group is nonsymmorphic with two U atoms in the primitive cell. This means that some group operations must be followed by nonprimitive translations and others lead to the interchange of the two uranium sites. The effect of group operations on $|\mu\rangle$ is well known, since they transform as Γ_7, Γ_8, and Γ_9. This has been tabulated in Ref. 7, for instance, but the author rederived all relations used in the programs as a check. Finally, in the approximation used in this paper, Bloch's theorem will appear by factors of the form e^{ikR} in the matrix elements where R is the site position of a uranium ion. These cancel one another for the on-site case, but must be carefully kept track of for near-neighbor and next-near-neighbor terms.

The nature of the operator X will now be discussed. In the ^{3}He problem, it is taken to be the Pauli spin matrix. The simplest generalization for the current case is to replace σ by the pseudospin matrix τ, where τ has the same effect on k, PTk (or Pk,Tk) that σ has on up and down spinors (a normalization is used such that the determinant of the matrix is one). Note that P is the parity operator and T the time-reversal operator (P flips between sites relative to an inversion center; T flips between μ and $-\mu$). Further generalizations can be made by replacing τ by J, the total angular momentum operator (this is an approximation to a Hartree-Fock vertex). Further, if one is interested in quadrupolar interactions, thought by some to be of fundamental significance in the heavy-fermion problem,[8] then one can in turn replace J by

$$O_2^0=[3J_z^2-J(J+1)]/\sqrt{3} ,$$
$$O_2^{1,a}=[J_z(J_+ +J_-)+(J_+ +J_-)J_z]/4 ,$$
$$O_2^{1,b}=[J_z(J_+ -J_-)+(J_+ -J_-)J_z]/(4i) ,$$
$$O_2^{2,a}=(J_+^2 +J_-^2)/4 ,$$

or

$$O_2^{2,b}=(J_+^2 -J_-^2)/(4i) .$$

To determine the appropriate symmetry of the gap function, one must project the pairing kernel onto each group representation. For UPt_3 these are Γ_1 through Γ_6, equivalent to the more common notation of $A_1, A_2, B_1, B_2, E_2, E_1$.[7,9] Moreover, a projection must also be done on odd and even parity. For even parity, the pair function is

$$|k,Tk\rangle-|PTk,Pk\rangle .$$

The odd-parity case involves a three-component vector. An assumption will be made in this case. Experimentally, it appears from neutron-scattering data[10] that the moments in UPt_3 are confined to the basal plane. If a similar effect occurs for the Cooper pair moments, then one expects only the "d_z" component of the odd-parity order parameter vector to be important. Such an order parameter is consistent with a recent analysis of H_{c2} anisotropy in UPt_3.[11] T_c depends only on d_z in the τ model if one assumes that $V^{xx}=V^{yy}$ with the rest of the $V^{ij}=0$. For the other models, all components play a role. It is assumed, though, that there is some term in the pairing Hamiltonian which also acts in these cases to project onto d_z.[12] For the relativistic case, d_z is

$$|k,Tk\rangle+|PTk,Pk\rangle .$$

To summarize, the following X_iX_j will be explored:

$$\tau_x\tau_x+\tau_y\tau_y,\quad J_xJ_x+J_yJ_y,\quad O_2^0O_2^0 ,$$
$$O_2^{1,a}O_2^{1,a}+O_2^{1,b}O_2^{1,b},\quad O_2^{2,a}O_2^{2,a}+O_2^{2,b}O_2^{2,b} .$$

The weak-coupling gap equation to be solved is

$$\Delta_k=\lambda^{-1}N\sum_{k'}V_{k,k'}\Delta_{k'} , \tag{3}$$

where

$$V_{k,k'}=\sum_{R,R',i,j}V_{RR'}^{ij}[\langle k|X_i|k'\rangle_R\langle Tk|X_j|Tk'\rangle_{R'} \mp\langle k|X_i|PTk'\rangle_R\langle Tk|X_j|Pk'\rangle_{R'}] , \tag{4}$$

N is the density of states, and λ^{-1} is the inverse coupling constant [equal to $\ln(1.13\omega_c/T_c)$ with T_c the transition temperature and ω_c some cutoff energy], with the upper sign for even parity and the lower one for d_z odd parity. The sum is carried out over all k vectors on the Fermi surface. For UPt_3, a 137 k vector mesh is used on the Fermi surface in the irreducible wedge (which in this case involves five bands), noting that there are 24 group operations. The k vectors are weighted by the appropriate density of states factors derived from a tetrahedron decomposition of the Brillouin zone. Equation (3) is solved by standard matrix methods, with λ being the largest number for which the equation is satisfied.

In Table I, the results of the above formalism for UPt_3 are summarized. In column 2, an on-site interaction of unit value is looked at.[13] The sign in each case is chosen so as to yield a repulsive interaction (i.e., opposite to that which would give a nodeless s-wave solution). This sign is positive for the τ, J, and O_2^2 cases, but is negative for the O_2^0 and O_2^1 cases. Despite the "repulsive" value of the sign, solutions are indeed found, due to the momentum dependence of the matrix elements. The solution with the largest coupling constant either has A_{1g} symmetry or is odd parity (for the τ case, the interaction is repulsive for all even-parity states). Addition of a repulsive momentum-independent constant to the gap equation did not suppress the A_{1g} solution since this solution already has a complicated nodal structure. In fact, the stability of the A_{1g} solution over all other even-parity solutions is most likely due to this complicated nodal structure, since the nodal structure of the other representations is fixed by symmetry and therefore not free to adjust itself to minimize the free energy.

In the final four columns, results for ferromagnetic (positive) and antiferromagnetic (negative) unit values of J (near neighbor, between planes) and JN (next near neighbor, in plane) interactions are tabulated. Again, the

TABLE I. Pairing coupling constants for various group representations for UPt_3. U represents a repulsive on-site interaction of unit value (the sign chosen opposite to that which would give a nodeless s-wave solution). N and NN represent near-neighbor and next-near-neighbor interactions of unit value with either a positive sign (F) or negative sign (AF). The interaction vertex is denoted by τ for $\tau_x\tau_x+\tau_y+\tau_y$, J for $J_xJ_x+J_yJ_y$, O_2^0 for $O_2^0O_2^0$, O_2^1 for $O_2^1O_2^1$, and O_2^2 for $O_2^2O_2^2$. Listed are the group representations with the largest coupling constants (with the coupling constant λ in parentheses).

Case	U	F-N	AF-N	F-NN	AF-NN
τ	B_{1u}(0.030)	E_{1u}(0.080)	A_{1g}(0.128)	B_{1u}(0.094)	A_{1g}(0.328)
	A_{2u}(0.019)	B_{1u}(0.072)	E_{2g}(0.094)	E_{1u}(0.092)	E_{2g}(0.140)
J	A_{1g}(0.058)	E_{1u}(0.188)	A_{1g}(0.209)	B_{1u}(0.280)	A_{1g}(0.728)
	B_{1u}(0.047)	B_{1u}(0.169)	E_{2g}(0.172)	E_{1u}(0.236)	E_{2g}(0.397)
				A_{1g}(0.207)	
O_2^0	A_{2u}(0.247)	A_{1g}(0.838)	A_{1g}(1.33)	A_{1g}(2.32)	A_{2u}(0.484)
	E_{1u}(0.171)	A_{2u}(0.699)	E_{2g}(0.600)	E_{1u}(0.635)	B_{1u}(0.334)
	E_{2g}(0.161)	E_{1u}(0.392)	E_{1u}(0.413)	B_{1u}(0.629)	A_{1g}(0.328)
					E_{1u}(0.323)
O_2^1	A_{1g}(0.157)	A_{1g}(0.438)	A_{1g}(0.361)	A_{1g}(1.60)	A_{1g}(0.547)
	B_{1u}(0.121)	A_{2u}(0.216)	E_{2g}(0.227)	E_{2g}(0.486)	E_{1u}(0.323)
	E_{2g}(0.102)	B_{1u}(0.215)	A_{2u}(0.186)		B_{1u}(0.272)
		E_{1u}(0.194)			
O_2^2	A_{1g}(0.145)	A_{1g}(0.374)	A_{1g}(0.236)	A_{1g}(0.381)	A_{1g}(1.05)
	B_{1u}(0.141)	E_{1g}(0.100)	E_{1u}(0.235)	E_{2g}(0.174)	E_{2g}(0.571)
			E_{2g}(0.166)	B_{1u}(0.163)	B_{1u}(0.392)

solution with the largest coupling constant either has A_{1g} symmetry or is odd parity. An interesting point to remark on is that for the τ and J cases, one finds that the ferromagnetic sign supports odd-parity solutions whereas the antiferromagnetic sign supports even parity (for the τ case, the ferromagnetic case is repulsive for all even-parity states and the antiferromagnetic case is repulsive for all odd-parity states). This is in accord with earlier wisdom on this subject[1] but did not occur for realistic calculations on UPt_3 which took the pairing kernel to be of the simple form $V(k-k')$.[2] This illustrates the crucial role band indices play when evaluating the pairing kernel (note the multiband nature of the problem is a direct consequence of the degeneracy of the f orbital and the multiple number of formula units per primitive cell). Another interesting point is that quadrupolar interactions prefer A_{1g}, regardless of the sign, in ten of twelve cases. A final note about Table I is that it is somewhat meaningless to compare the results for τ, J, and O against one another since each operator has a different physical origin and thus different matrix elements.

The most favored explanation for the large body of experimental data on UPt_3 is that the order parameter is from a two-dimensional group representation with line nodes orientated perpendicular to the c axis.[14] In the current context, this could be realized by E_{1g} or E_{2u}, the latter being consistent with the H_{c2} anisotropy analysis.[11] Neither state is favored by any of the pair interactions analyzed in the current paper [the same was found for earlier work based on the simple form $V(k-k')$].[2] The most favored solution is A_{1g}, probably due to the lack of symmetry restrictions on its nodal structure. This casts doubts on any even-parity interpretation of the data. As for odd-parity solutions, the two-dimensional E_{1u} representation is found in several cases. This solution has point nodes. On the other hand, arguments have been put forth that the observed line node structure in UPt_3 may not be due to the symmetry of the order parameter, but rather to gaplessness due to the normal-state self-energy.[15] More work will certainly be necessary before the actual form of the order parameter is unambiguously determined.

An alternate explanation of the data would be a near degeneracy of two different representations.[16,17] This picture is possibly supported by the current work, which indicates several such cases which involve one single-dimensional and one two-dimensional representation. For instance, for the τ case with U, J, and $JN>0$, B_{1u} and E_{1u} remain nearly degenerate over a large region of parameter space.

The author concludes with some remarks on strong coupling effects. The author has performed calculations on UPt_3 including the full momentum and frequency dependence of the pairing interaction and treating all k vectors and f bands (instead of restricting to the Fermi surface) as recently advocated by Monthoux and Pines,[5] using the simple $V(k-k')$ model. Although this treatment leads to a renormalization of the value of T_c, no change in the ordering of the solutions with respect to coupling constant was seen (this is a consequence of the fact that the frequency dependence of the self-energy is much stronger than the momentum dependence). For

that reason, one would suspect that strong-coupling effects would not alter the ordering of solutions seen here, although it is conceivable that variation of the character of the band wave functions off the Fermi surface could alter the results. Such effects could be tested in the future if the effort seems warranted.

As a final thought, the author would like to emphasize the generality of the above method. Currently, this has been applied to a phenomenological and very simple interaction vertex for UPt_3 as an example. Once a more microscopic theory is available,[18] then the resulting calculation for properly testing such a theory should follow a similar formalism as expounded in this paper.

In summary, a method is derived for evaluating the pairing kernel in heavy-fermion superconductors utilizing matrix elements determined from relativistic band-structure calculations. As an example, several models for the pairing interaction for UPt_3 were considered, yielding a rich variety of solutions. Further work, both experimental and theoretical, will be necessary before it can be determined what relevance the determined solutions have to the experimental situation.

This work was supported by U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38.

[1]K. Miyake, S. Schmitt-Rink, and C. M. Varma, Phys. Rev. B **34**, 6554 (1986); D. J. Scalapino, E. Loh, Jr., and J. E. Hirsch, *ibid.* **34**, 8190 (1986).

[2]M. R. Norman, Phys. Rev. Lett. **59**, 232 (1987); Phys. Rev. B **37**, 4987 (1988); **39**, 7305 (1989); **41**, 170 (1990); **43**, 6121 (1991).

[3]W. Putikka and R. Joynt, Phys. Rev. B **37**, 2372 (1988); **39**, 701 (1989).

[4]B. Welslau and N. Grewe, Ann. Phys. **1**, 214 (1992).

[5]P. Monthoux, A. V. Balatsky, and D. Pines, Phys. Rev. Lett. **67**, 3448 (1991); A. J. Millis, Phys. Rev. B **45**, 13 047 (1992); P. Monthoux and D. Pines, Phys. Rev. Lett. **69**, 961 (1992); R. J. Radtke, S. Ullah, K. Levin, and M. R. Norman, Phys. Rev. B **46**, 11 975 (1992).

[6]P. W. Anderson, Phys. Rev. B **30**, 4000 (1984); K. Ueda and T. M. Rice, *ibid.* **31**, 7114 (1985).

[7]J. Appel and P. Hertel, Phys. Rev. B **35**, 155 (1987).

[8]D. L. Cox, Phys. Rev. Lett. **59**, 1240 (1987).

[9]G. E. Volovik and L. P. Gor'kov, Zh. Eksp. Teor. Fiz. **88**, 1412 (1985) [Sov. Phys. JETP **61**, 843 (1985)].

[10]G. Aeppli, E. Bucher, A. I. Goldman, G. Shirane, C. Broholm, and J. K. Kjems, J. Magn. Magn. Mater. **76-77**, 385 (1988).

[11]C. H. Choi and J. A. Sauls, Phys. Rev. Lett. **66**, 484 (1991).

[12]One cautionary note is that the d vector in this formalism resides in pseudospin space which has a nontrivial relation to real space. Thus, the projection onto d_z may not be totally appropriate. Including the other two components of the odd-parity state, $|k,Pk\rangle$ and $|PTk,Tk\rangle$, is relatively straightforward, but leads to a complicated matrix equation.

[13]A more sophisticated treatment would consider higher multipolar terms for the on-site interaction based on a crystal-field-type model.

[14]R. Joynt, J. Magn. Magn. Mater. **108**, 31 (1992) and references therein.

[15]M. R. Norman, Physica C **194**, 203 (1992).

[16]R. Joynt, V. P. Mineev, G. E. Volovik, and M. E. Zhitomirsky, Phys. Rev. B **42**, 2014 (1990).

[17]M. Ozaki and K. Machida, J. Phys. Soc. Jpn. **61**, 1277 (1992).

[18]An example would be a realistic calculation of $\chi(r,r')$ with the proper dependence on band and pseudospin index. A more general approach would be to fit the particle-hole vertex to mass renormalization factors extracted from deHaas-vanAlphen data, and then use this information to construct the particle-particle vertex. This is analogous to what has been done in ^{3}He, where the pair vertex was fit using the experimental Landau parameters.