Superconductivity in Almost-Localized Fermi Liquids: Application to Heavy-Fermion Compounds

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We consider the Landau theory of an almost-localized Fermi liquid with a very large m^*/m ratio. We argue that this system has a superconducting transition to a p-wave state. We estimate T_c, calculate the specific-heat discontinuities at T_c, and discuss phase stability. Using available experimental data for γ and k_F, we find that our results for this model are in good agreement with the known experimental values for heavy-fermion compounds.

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The discovery of superconductivity in heavy-fermion compounds[1-4] such as UBe_{13}, $CeCu_2Si_2$, UPt_3, and U_6Fe has generated a great deal of excitement. These compounds exhibit values of γ (defined by the low-temperature behavior of the specific heat, $C_v=\gamma T$) which are larger, by 2 or 3 orders of magnitude, than typical metallic values. This translates into astonishingly large effective masses. The first two of the above-named compounds clearly exhibit Kondo behavior in their electrical resistivity, while for the other two the situation is less clear. It is thought that superconductivity in these compounds is not due to the usual phonon mechanism but it is caused by Landau Fermi-liquid effects,[5-7] that is, spin-fluctuation effects involving the heavy electrons. This brings up immediately the analogy with ^{3}He and the possibility of exotic pairing states.[5,6] In the case of UBe_{13}, this seems to have been experimentally confirmed[7]: The specific heat below T_c decreases at T^3 which is consistent with an Anderson-Brinkman-Morel[8] type of p-wave superconducting state.

It is natural to attempt[5,7] to put these ideas on a more quantitative footing by using the standard paramagnon model[9] of spin fluctuations as used for ^{3}He. This is, however, impossible for those compounds: Within the paramagnon model the effective mass diverges very slowly as the spin-fluctuation parameter $\bar{I}$ approaches unity. At $1-\bar{I}=10^{-4}$, the effective-mass ratio m^*/m is less than 40.[10] For this value of $\bar{I}$, the paramagnetic susceptibility χ in the normal state would be (according to the paramagnon model) at least several hundred times larger than for free fermions of mass m^*. This is in complete contradiction with the behavior of heavy-fermion compounds: While the paramagnetic part of the susceptibility is difficult to extract experimentally, it is very clear from the experimental evidence that χ/γ does not exceed its free-particle value by any large amount. Further, in the limit $\bar{I}=1$ the pairing-transition temperature vanishes[10,11] within the paramagnon model: An extremely large effective mass would imply the disappearance of superconductivity. These discrepancies are simply too important to be ignored. A less obvious quantitative discrepancy is found also[7] when attempting to fit the value $\Delta C/C_N$ of the specific-heat discontinuity at T_c to the paramagnon theory of Ref. 12, which contains (besides $\bar{I}$) an additional parameter δ. The value $\delta=1.0$ which fits experiment[7] would correspond to unphysically large values of the enhancement region in k space.

There are, however, alternatives to the paramagnon model which have been developed to treat Landau Fermi liquids in the context of ^{3}He. Liquid ^{3}He can be viewed[13] as being close to localization, rather than close to ferromagnetism.[14] This point of view exploits the ideas of Gutzwiller[15] and its application to ^{3}He has been recently reviewed.[16] There has been speculation[17] that these methods may prove fruitful in the heavy-fermion problem. The main reason for this is that within this approach the susceptibility enhancement factor does not diverge when the effective mass is very large, but it tends to a limiting value of 4 (the experimental values which are quoted in the literature for this enhancement factor are somewhat smaller than this, which we believe is due to spin-orbit and diamagnetic effects not included in our model). This is a strong indication that the quasilocalization point of view may prove quantitatively more appropriate than the paramagnon model to the study of heavy-fermion superconductivity.[18]

In this Letter we will consider the Landau theory of an almost-localized isotropic Fermi liquid of spin-$\frac{1}{2}$ particles, in the limit when the effective-mass enhancement is very large. The values of several of the Landau parameters can easily be determined in this case. We will then show that such a liquid undergoes a transition to a state with

$l=1$ pairing. We will also calculate the specific-heat discontinuities at the transition and determine which phase[8] is stable below it. The results account quite well, in respect to the properties we discuss, for the behavior of the heavy-fermion compounds. We are assuming, of course, that an important part of the very large effective mass in these compounds is of many-body origin, rather than a band-structure effect.

Since several of the Landau parameters F_l diverge at the localization transition, it is more convenient to work with the Landau amplitudes $A_l^{s,a}=F_l^{s,a}[1+F_l^{s,a}/(2l+1)]^{-1}$. When the effective-mass ratio is so large, we have $A_1^s=3$. Further, in a system close to localization, we have[13] $A_0^s=1$.[19] It follows from Refs. 13 and 16 that $A_0^a=-3$ as explained above. The remaining parameters are not known. We will assume that $A_l^{s,a}=0$ for $l\geq 2$ and attempt to compensate for any error thus introduced by setting $A_1^a=-1$, which ensures that the forward-scattering sum rule $\sum_l(A_l^s+A_l^a)=0$ is properly satisfied.

Within this formulation of Fermi-liquid theory, the coupling constant for pairing in the s or p partial waves can be calculated in terms of the Landau parameters with the method of Patton and Zaringhalam.[20] For $l=1$ the result is

$$\lambda=-\sum_l(-1)^l(A_l^s+A_l^a)/12. \quad (1)$$

Substituting in Eq. (1) our values of the Landau amplitudes, we find $\lambda=\frac{1}{3}$. The sign corresponds to an attractive interaction. A similar calculation for the s wave yields a repulsive λ. Thus, the possibility of s-type BCS pairing is ruled out. The system is, however, unstable to the formation of p-wave Cooper pairs. The critical temperature for this transition may be written[20]

$$T_c=2T^*\exp(-1/\lambda), \quad (2)$$

where T^*[21] is a characteristic temperature corresponding to the cutoff energy. In ^{3}He T^* is of the order of the spin-fluctuation temperature, $T_{sf}\simeq 0.1T_F$.[9] In heavy-fermion compounds T^* arises naturally as the width of the narrow feature in the density of states at the Fermi level. It can be experimentally identified either as the degeneracy temperature in the specific heat versus temperature curve or as the Kondo temperature in those compounds (e.g., $CeCu_2Si_2$ and UBe_{13}) with a Kondo anomaly. For $CeCu_2Si_2$ and UBe_{13} both methods give the same result: $T^*\simeq 10$ K.[1,2] For UPt_3 (which shows no Kondo feature in the resistivity ρ) one also has $T^*\simeq 10$ K from the temperature dependence of the specific heat.[3] Therefore, we obtain for these three compounds $T_c\simeq 1$ K, in very satisfactory agreement with the experiment, considering the simplicity of our approach. The above argument explains also why the T_c of U_6Fe (for which no determination of T^* is available) is somewhat larger than for other uranium compounds: Uranium atoms in U_6Fe are packed closer together and the increased overlap of the f shells causes a broadening of the band, thereby increasing T^* and therefore T_c. This is consistent with γ being, in U_6Fe, smaller than in UBe_{13} and UPt_3. The above considerations indicate $T^*\simeq 40$ K for this compound.

Our picture of the quasilocalized electron liquid implies therefore that the superconducting state is of the p type. It also implies that λ varies very little from compound to compound, provided that m^*/m is very large, and thus that T_c scales with T^*. If pressure is applied to the system, the overlap between f orbitals increases, and this results in an increase in T^*. As long as the pressure is not too large (so that m^*/m remains much larger than unity), T_c will increase with pressure, within our model, the increase being proportional to the corresponding increase in T^*. This relation between T_c and T^* is experimentally verifiable.[22]

The complexity of the order parameter in p-type superconductivity allows for various different phases[8] as possible candidates for the equilibrium state below T_c. In the weak-coupling limit the isotropic Balian-Werthamer state is always stable[8] but the size of λ which we have found indicates that we should consider strong-coupling corrections. The effect of these corrections is to change the the coefficients β_i $(i=1,\ldots,5)$, which appear in the Ginzburg-Landau free energy,[8] from their weak-coupling values β_i^{BCS}. From the values of β_i one can determine not only which is the stable phase, but also the specific-heat discontinuity at the transition, $\Delta C/C_N$.

The change $\Delta\beta_i=\beta_i-\beta_i^{BCS}$ can be calculated with the method of Rainer and Serene.[23] Within their method one finds $\Delta\beta_i=\eta B_i$, where the B_i can be expressed, with use of (as in Ref. 20) the s-p approximation for the scattering amplitudes, as functions of the Landau amplitudes only, and $\eta=\epsilon\pi^2k_BT_c/7\zeta(3)\hbar k_Fv_F$.

To calculate η, therefore, we need to know both the Fermi wave vector k_F and the Fermi velocity v_F. In order to obtain this information we need, in addition to the experimental values of γ and the mass density, the effective number of conduction electrons per molecule, Z. This must be extracted

TABLE I. The first three columns are the values of the quantities γ, T_c, and k_F, extracted from experiment and used in our calculation of the specific-heat discontinuities $\Delta C/C_N$ (see text). The experimental results for $\Delta C/C_N$ are given next. The theoretical values of $\Delta C/C_N$ and stable phases are the results, within our model, for a system having the values of γ, T_c, and k_F quoted here for each compound. All experimental data that we use can be found in Refs. 1–4, 7, and 26.

Compound	γ (J/K^2 mole)	T_c (K)	k_F (Å^{-1})	$\Delta C/C_N$, expt.	$\Delta C/C_N$, theor.	Stable phase
$CeCu_2Si_2$	~ 1	0.64	1.7	~ 1	1.7	B
UBe_{13}	1.1	0.9	1.4	2.5	2.3	A
UPt_3	0.45	0.54	1.6 (1.08)	>1	1.6 (2.0)	B (B)
U_6Fe	0.155	3.8	1.6	2.1	1.8	B

from a separate experiment. In the case of UBe_{13}, $Z=11$ has been obtained[1] from the Kondo maximum in the resistivity. The resulting value of k_F is $k_F=1.4$ Å^{-1}.[24] For $CeCu_2Si_2$ a value of Z equivalent to $k_F=1.7$ Å^{-1} is reported from critical-field slope measurements.[2] We have verified that this value is consistent with that obtained from the maximum electrical resistivity ρ_m. No published experimental information about Z is available for the other two compounds. For the nonsuperconducting heavy-fermion compound $CeAl_3$, one finds $k_F=1.6$ Å^{-1} from ρ_m.[25] The above values of k_F are typical for metals, and consequently we have taken $k_F \simeq 1.6$ Å^{-1} for UPt_3 and U_6Fe in our calculations.[26] Once k_F is specified, the $\Delta\beta_i$'s can be calculated with use of our values for the $A_i^{s,a}$, and the results can be analyzed[23] to determine the equilibrium state and $\Delta C/C_N$. Using the experimental values of γ and T_c,[1-4] and the values of k_F quoted above, we find within the s-p approximation that the Balian-Werthamer state is stable for the values of these quantities corresponding to $CeCu_2Si_2$, U_6Fe, and UPt_3. In the case of U_6Fe this would not hold if k_F were found experimentally to be much smaller than 1.6 Å^{-1}. Then, the A phase (axial or Anderson-Brinkman-Morel state[8]) would be stable. For the values of γ, T_c and k_F corresponding to UBe_{13}, we find that the A phase is stable. This result is in complete agreement with the experimental fact[7] that the specific heat for this compound goes as T^3 at low T.

Our results for the specific-heat discontinuities at the transition are summarized in Table I. The agreement with the available experimental data is remarkably good, considering that our model is indeed very simple. Our results also exhibit the trend found in the experimental values of $\Delta C/C_N$ when one moves from one compound to another.

In conclusion, we have considered in this Letter a quasilocalized Fermi liquid in the limit where the mass-enhancement factor is very large. Using the Landau parameters obtained for this model, we have shown that it exhibits triplet pairing; we have found values for T_c and $\Delta C/C_N$ and discussed phase stability. It is clear that this model does not include many of the features necessary for a detailed understanding of heavy-fermion compounds (anisotropy, spin-orbit coupling, impurities, competition with magnetism, etc.) and that additional work is needed in these directions. We do not claim to have shown conclusively that our model is indeed applicable to heavy-fermion compounds. Nevertheless, the results of our model are extremely satisfactory, from the numerical standpoint, and very suggestive as to what is the actual physics of these compounds.

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