Signatures of pairing mechanisms and order parameters in ferromagnetic superconductors

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Two predictions are made for properties of the ferromagnetic superconductors discovered recently. The first one is that spin-triplet, p-wave pairing in such materials will give the magnons a mass inversely proportional to the square of the magnetization. The second one is based on a specific mechanism for p-wave pairing, and predicts that the observed broad anomaly in the specific heat of URhGe will be resolved into a split transition with increasing sample quality. These predictions will help discriminate between different possible mechanisms for ferromagnetic superconductivity.

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Recently there has been a considerable amount of work on what is believed to be true coexistence between ferromagnetism and superconductivity. In UGe$_2$ [1], ZrZn$_2$ [2], and URhGe [3], superconductivity has been observed at very low temperatures inside the ferromagnetic phase.

There have been several theoretical suggestions for the cause of this exotic superconductivity. Early theories proposed magnetically mediated spin-triplet superconductivity in ZrZn$_2$ [2], and recent band-structure calculations have concluded that this is a possibility in these materials [3]., and Machida and Ohmi [7] have give arguments for the pairing to be of p-wave, non-unitary spin-triplet nature. On the other hand, a conventional phonon mechanism has been proposed in Ref. [8], and Ref. [8] has suggested that the superconductivity is due to coupled CDW and SDW fluctuations. It is not obvious from these considerations why the superconductivity is observed only in the ferromagnetic phase. To explain this, Sandeman et al. have proposed a density-of-states effect, that exists only in the ferromagnetic phase, as the source of the superconductivity in UGe$_2$ [10]. Kirkpatrick et. al. [11, 12] have proposed an explanation for the observed phase diagram that is based on an enhancement of the longitudinal spin susceptibility in the ferromagnetic phase by magnons, or magnetic Goldstone modes.

In this Letter we predict two observable effects that allow to partially discriminate between these various theoretical mechanisms. First, we show that if the order parameter (OP) is of non-unitary spin-triplet type, the ferromagnetic magnons, or transverse spin fluctuations, become effectively massive in the superconducting phase, with a mass that is inversely proportional to the square of the magnetization. This prediction depends only on the symmetry of the OP, and is independent of the mechanism for superconductivity. The second prediction is based on our previously suggested mechanism for superconductivity in these systems [11, 12]. It predicts that the specific heat anomaly observed in URhGe around the superconducting transition temperature $T_c$ actually consist of two distinct features that will be resolved as the sample quality increases. These two features correspond to the pairing of electron spins parallel and antiparallel, respectively, to the direction of the magnetization. In what follows, we will first give our detailed results and simple intuitive explanations, and then we will sketch their technical derivation [13].

General symmetry principles [14] show that, in the presence of a nonvanishing magnetization in $z$-direction, the magnetic susceptibility tensor has the structure

$$\chi = \begin{pmatrix} \chi_{T,+} & \chi_{T,-} & 0 \\ -\chi_{T,-} & \chi_{T,+} & 0 \\ 0 & 0 & \chi_L \end{pmatrix},$$

with $\chi_{T,\pm}$ and $\chi_L$ the transverse and longitudinal magnetic susceptibilities, respectively. In the absence of superconductivity, $\chi_{T,\pm}$ is massless, since it is the magnetic Goldstone mode [14]. Spin-triplet superconductivity explicitly breaks the spin rotation symmetry and leads to a mass in $\chi_{T,\pm}$. For small frequencies $\Omega$ and wavevectors $k$ we find

$$\chi_{T,\pm}(k, i\Omega) = \left[ \frac{a_{\pm} \delta}{i\Omega + \delta b (k^2 + \mu^2)} \right] \pm (\delta \rightarrow -\delta).$$

Here $\Omega = \Omega/4\epsilon_F$ and $k = k/2k_F$ are the frequency and wavevector made dimensionless by means of the Fermi energy $\epsilon_F$ and the Fermi wavenumber $k_F$, respectively. $a_+$ and $b$ are real constants on the order of unity, and $a_-$ is imaginary with a modulus on the order of unity. $\delta = \delta/4\epsilon_F$, with $\delta$ the Stoner splitting of the Fermi surface, which is proportional to the magnetization $m$. In a phase where only up-spin electrons are paired, with $\Delta_T$ the up-spin superconducting energy gap, we find for the mass

$$\mu^2 = (\Delta_T/2\delta)^2 f(\delta/2T),$$

where $f(z)$ is a function of the form $f(z) = z^p (1 - z^q)$.
heat discontinuity ∆

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FIG. 1: Circles denote the experimental\( T \) specific heat co-
efficient of URhGe [3]. The solid lines show the theoretical expectation if only half of the electrons pair (a), and if there are two transitions within 5% of one another (b). See the text for additional information.

where

\[
f(x) = \int_0^\infty \frac{dy}{y(1-y^2)} \, \tanh(yx) .
\] (3b)

Asymptotic analysis yields \( f(x \to \infty) = \ln x \), and \( f(x \to 0) = 0.85 \ldots x^2 \). Such a mass in the transverse magnetic susceptibility is in principle observable by neutron scattering. An estimate using \( \Delta_{1} \approx 2T_{c1} = 1 \text{K} \), \( \delta = \epsilon_{F}/2 = 10^{4} \text{K} \), and \( k_{F} = 1 \text{Å}^{-1} \), yields \( 2k_{F}\mu = 3 \times 10^{-4} \text{Å}^{-1} \). This is roughly a factor of 30 smaller than the current lower wavenumber limit for inelastic neutron scattering [10]. Since \( \mu \) depends roughly linearly on \( \Delta_{1} \), a higher value of \( T_{c} \) would help push \( \mu \) into an observable regime.

Our second prediction concerns the detailed structure of the specific heat anomaly observed in URhGe [3]. The experiment shows a broad peak with a maximum close to the resistive \( T_{c} \), which is evidence for true bulk superconductivity. However, for low temperatures the specific heat coefficient does not go to zero, but rather approaches a finite value that is roughly half its value in the normal phase, see Fig. 1. Ref. 3 has interpreted this behavior as an indication for the pairing being restricted to one spin projection, while the electrons with the opposite spin projection retain a Fermi surface down to the lowest temperatures measured. With a relative specific heat discontinuity \( \Delta C/C \) adjusted to fit the data, and a temperature dependence below \( T_{c} \) given by \( C(T)/T \propto T^{1} \) [17], this suggests that, with improving sample quality, the data should approach the solid curve shown in Fig. 1(a). This is not the only possible interpretation. We have extended our previous calculation of the superconducting \( T_{c} \) to include both up-spin and down-spin pairing. For the superconducting gap, which is a \( 2 \times 2 \) matrix \( \Delta \) in spin space, we assume [18]

\[
\hat{\Delta}(\mathbf{k}) = \begin{pmatrix}
\Delta_{1}\hat{k}_{z} & 0 \\
0 & -\Delta_{1}\hat{k}_{z}
\end{pmatrix},
\] (4)

with \( \hat{k} \) a unit wavevector. This is the OP of the \( A_{2} \) phase in He [10]. For generic parameter values we find that there is only a small difference between the values of \( T_{c1} \) and \( T_{c1} \) in the ferromagnetic phase, on the order of 5-10%. This is comparable to the difference Fay and Appel [4] found within a simpler theory that could not describe the strong enhancement of \( T_{c} \) in the ferromagnetic phase. Consequently, one expects that in a perfect sample the specific heat data would qualitative look as shown in Fig. 1(b). In this interpretation, the broad peak hides a split transition, and the low-temperature tail of the specific heat coefficient is due to non-superconducting parts of the sample. (See, however, Ref. 20.) It is interesting that such a split transition has been observed in the heavy-fermion superconductor UPt [21, 22] (which is not ferromagnetic, and where the splitting has a different physical origin), but only after a long period of improving sample qualities.

We now turn to the origin of these results, starting with the mass in the magnon. Any spin-triplet OP can be represented as a vector \( \phi \) in spin space that is isomorphic to the \( 2 \times 2 \) spin matrix representation [10]. For the OP given in Eq. 4, this vector is proportional to

\[
\phi = (\Delta_{1} - \Delta_{1}, i(\Delta_{1} + \Delta_{1}), 0) .
\]

We see that \( \phi^{*} \) is not parallel to \( \phi \). This is an example of a “non-unitary” OP, and Ref. 7 has argued that any OP in ferromagnetic superconductors is likely of this general type. In a Landau theory, the lowest order term in the free energy density that couples the magnetization vector \( \mathbf{M} = (0,0,m) \) and the superconducting OP that is allowed by symmetry is thus of the form

\[
f_{\text{coupling}} = icM \cdot (\phi \times \phi^{*}) ,
\] (5)

with \( c \) a real constant. This shows that the presence of a nonzero superconducting OP, \( \Delta_{1} \) is equivalent to an effective magnetic field \( \hbar a \hbar \propto \Delta_{1}^{2} \). General symmetry considerations [23] show that the transverse magnetic susceptibility must then have a mass that is given by \( \chi_{\Delta_{1}^{-1}}(0,0) = \hbar a \hbar /m \). This explains the existence of a mass, and the dependence of \( \mu^{2} \), Eq. 3(a), on \( \Delta_{1} \).

For an understanding of the remaining structure of \( \mu^{2} \), as well as of our second prediction, we return to our previous work on the superconducting transition of the up-spin electrons [12]. As in that paper, we consider a model of free electrons with a static, point-like spin-triplet interaction with amplitude \( \Gamma \). For superconducting OP fields we choose, \( F_{\uparrow}(x,y) = \psi_{\uparrow}(x)\psi_{\uparrow}(y) \) and \( F_{\downarrow}(x,y) = \psi_{\downarrow}(x)\psi_{\downarrow}(y) \), with \( \psi_{\sigma}(x) \) an electronic field with spin index \( \sigma \) and space-time index \( x \). The OPs, i.e., the expectation values \( \langle F_{\sigma}(x,y) \rangle = F_{\sigma}(x-y) \), are the anomalous Green functions.

Using a technique similar to that employed in Ref. 12, we have derived coupled equations of motion for the \( F_{\sigma} \), the normal Green functions \( G_{\sigma} \), and the magnetization
find the following Eliashberg-type equations for the gap functions $\Delta_\sigma$ and the normal self-energies $\Sigma_\sigma$,

$$\Delta_\sigma(k) = \frac{1}{\Gamma_1} \int q \chi_L(k-q) \Delta_\sigma(q)/d_\sigma(q), \quad (6a)$$

$$\Sigma_\sigma(k) = \frac{1}{\Gamma_1} \int q \chi_L(k-q) G_\sigma^{-1}(q)/d_\sigma(q) + 2\Gamma_1 \int q [\chi_{T,+}(k-q) + i\chi_{T,-}(k-q)] G_\sigma^{-1}(q)/d_-\sigma(q), \quad (6b)$$

with $\sigma = \uparrow, \downarrow \equiv +, -$. Here we use a wavevector-frequency four-vector notation $k = (k, \omega_n)$, and $\int_q = T \sum_\alpha (1/V) \sum_q$. We have introduced $d_\sigma(k) = G_\sigma^{-1}(q) = \Delta_\sigma(k)\Delta_\sigma^*(k)$, and the $G_\sigma^{-1}(k) = i\omega_n - (k^2/2m_n - \mu - \sigma\delta - \Sigma_\sigma(k))$ are the inverse 'normal' Green functions, where $\delta = \Gamma_1m$ is the Stoner energy. The magnetic equation of state to one-loop order is given by,

$$m = \frac{T}{2V} \text{Tr} (\gamma_3 G) + \frac{\Gamma_1}{2} \int dx \, dy \sum_{jk} \chi_{jk}(x-y) \times \text{tr} \left[ \gamma_3 G(x-y) \gamma_j G(x-y) \gamma_k G(y) \right]. \quad (7)$$

It is easy to show that the magnetic susceptibility $\chi$ that serves as the effective potential in these equations is proportional to the physical spin susceptibility, Eq. (11).

In a Gaussian approximation $^{[24]}$ we find

$$\chi_{ij}^{-1}(x-y) = \delta_{ij} \delta(x-y) + \frac{\Gamma_1}{2} \text{tr} \left[ G(y-x) \gamma_i G(x-y) \gamma_j G(y) \right]. \quad (8)$$

In these equations, the $(\gamma_0, \gamma) = (\sigma_3 \otimes \sigma_0, \sigma_3 \otimes \sigma_1, \sigma_0 \otimes \sigma_2, \sigma_3 \otimes \sigma_3)$ are generalized Nambu matrices, and the $G$ are $4 \times 4$-matrix Green functions that represent the normal and anomalous Green functions in this basis. $\text{Tr}$ denotes a trace over all degrees of freedom, and $\text{tr}$ denotes a trace over the discrete degrees of freedom.

An evaluation of Eq. (8) leads to a transverse susceptibility tensor that has the form of Eq. (2), with $a_+ = 1$, $a_- = -i$, $b = 2/3$, and $\mu^2$ as given in Eq. (3a). The general Landau theory argument given above, see Eq. (5), shows that this form of $\chi$ is generic, and not an artifact of our Gaussian approximation. (See, however, Ref. [12].)

The explicit calculation also shows that the prefactor of the $\Delta_T^2$ in the mass is a singular function of the magnetization and the temperature. This implies that a strict Landau expansion in powers of both the magnetic and the superconducting OPs is singular, since the coefficients of the Landau function do not exist. This breakdown of the Landau expansion is due to the soft particle-hole excitations in an itinerant electron system, and is analogous to an effect that has been discussed for quantum ferromagnets in the absence of superconductivity $^{[25]}$.

Our second prediction hinges on a solution of the gap equations, Eqs. (10), which require the magnetic susceptibility tensor $\chi$ as an input. A complete numerical solution of these strong-coupling equations would be very difficult, and does not seem worthwhile in the absence of detailed experimental information about $\chi$. In Ref. [12] we emphasized that the relative values of $T_{c1}$ in the paramagnetic and ferromagnetic phases, respectively, can be obtained within a simple McMillan-type approximation, even though the absolute values obtained by this method are probably not reliable. Here we adopt the same philosophy, with the aim being to compute the relative magnitudes of both $T_{c1}$ and $T_{c2}$. In the paramagnetic phase this is straightforward, using the Gaussian approximation, Eq. (3), for the magnetic susceptibility. We find results for $T_{c1}$ and $T_{c2}$ that are comparable to those obtained by Fay and Appel $^{[4]}$, i.e., a ratio $T_{c1}/T_{c2}$ that is, for generic parameter values, on the order of 1.1.

In the ferromagnetic phase we need to take into account the effect discussed in detail in Ref. [12], namely an enhancement of the longitudinal susceptibility due to a coupling of the latter to the ferromagnetic Goldstone modes $^{[26]}$. This requires a calculation of $\chi$ to one-loop order, beyond the Gaussian approximation given in Eq. (3). For $T_{c1}$ this calculation has been performed in Ref. [12]. For $T_{c2}$ there is the additional complication that inside the superconducting phase, $\chi$ depends on the up-spin gap $\Delta_T$. This introduces a feedback effect that is characteristic of any purely electronic mechanism for superconductivity. Since the overall effect of this feedback is rather small (see below), we can approximate the temperature dependent $\Delta_T$ in $\chi$ by $2T_{c1}$. We have found that $T_{c2}$, like its $T_{c1}$ counterpart, is enhanced over its value in the paramagnetic phase by a factor of 50-100, and for generic parameter values it is within about 10% of $T_{c1}$.

For the specific heat this leads to the prediction shown qualitatively in Fig. 1, namely, two transitions in close proximity. Our calculations therefore suggest that the specific heat peak measured in URhGe actually consist of two unresolved peaks. We note that the width of the experimentally observed peak easily encompasses both mean-field discontinuities. In contrast to our first prediction, the second one is specific to the pairing mechanism we have considered. If experiments on cleaner samples should show no indications of two closely spaced transitions, that would be a strong argument against the spin-fluctuation induced pairing we have considered.

We finally discuss our second result in some more detail. The salient point of our mechanism for a strongly enhanced superconducting $T_c$ in the ferromagnetic phase is the coupling of the massless (on the superconducting boundary, where $\Delta_T = 0$) transverse spin susceptibility to $\chi_L$ via mode coupling effects. Because $\chi_L$ serves as the pairing potential for the $\Delta_T$ ordering, this suggest that in the superconducting phase the pairing potential decreases, since $\chi_T$ decreases for all wavenumber and fre-
frequencies with increasing $\Delta_{\uparrow}$. For spin-triplet superconductivity in a magnetic field one generically expects an ordering of the spin down electrons, $\Delta_{\downarrow} \neq 0$, at some temperature $T_{c\downarrow} < T_{c\uparrow}$. Since the pairing potential for $\Delta_{\downarrow}$ is also $\chi_{\downarrow}$, this implies there is a weaker tendency for the onset of $\Delta_{\downarrow}$ given $\Delta_{\uparrow} \neq 0$, than there is for the onset of $\Delta_{\uparrow}$ ordering. Another general mechanism decreasing $T_{c\downarrow}$ compared to $T_{c\uparrow}$ is that the density of states at the Fermi surface for the down-spin electrons decreases with increasing magnetization. Naively, one therefore expects that for small magnetization the cutoff in Eq. (2), which is proportional to $\delta^{-2}$, greatly suppresses the $\Delta_{\downarrow}$ ordering, while for large $\delta$ the density of state effect should decrease it. This would result in, for example, a specific heat discontinuity at $T_{c\uparrow}$, and a substantial residual specific heat coefficient at lower $T$, since the down-spin electron would not undergo a superconducting transition until perhaps immeasurably low temperatures. This is certainly consistent with the experimental results in the URhGe system, and it is the interpretation given in Ref. 3. However, our detailed numerical work suggests that this is not the generic situation for the pairing mechanism we have considered. Clearly, with improving sample quality that will sharpen the feature in the specific heat, this question can be decided experimentally.

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[1] S. S. Saxena, P. Agarwal, K. Ahilan, F. M. Grosche, R. K. W. Haselwimmer, M. J. Steiner, E. Pugh, I. R. Walker, S. R. Julian, P. Monthoux, et al., Nature 406, 587 (2000).
[2] C. Pfleiderer, M. Uhlarz, S. M. Hayden, R. Vollmer, H. von Löhneysen, N. R. Bernhoeft, and G. G. Lonzarich, Nature 413, 58 (2001).
[3] D. Aoki, A. Huxley, E. Ressouche, D. Braithwaite, J. Floquet, J. P. Brison, E. Lhotel, and C. Paulsen, Nature 413, 613 (2001).
[4] D. Fay and J. Appel, Phys. Rev. B 22, 3173 (1980).
[5] A. Shick and W. Pickett, Phys. Rev. Lett. 86, 300 (2001).
[6] G. Santi, S. B. Dugdale, and T. Jarlborg, Phys. Rev. Lett. 87, 247004 (2001).
[7] K. Machida and T. Ohmi, Phys. Rev. Lett. 86, 850 (2001).
[8] H. Shimahara and M. Kohmoto, Europhys. Lett. 57, 247 (2002).
[9] S. Watanabe and K. Miyake, J. Phys. Soc. Jpn. 71, 2489 (2002).
[10] K. Sandeman, G. Lonzarich, and A. Schofield, Phys. Rev. Lett. 90, 167005 (2003).
[11] T. R. Kirkpatrick, D. Belitz, T. Vojta, and R. Narayanam, Phys. Rev. Lett. 87, 127003 (2001).
[12] T. R. Kirkpatrick and D. Belitz, Phys. Rev. B 67, 024515 (2003).
[13] D. Belitz and T. R. Kirkpatrick, unpublished results (2003).
[14] D. Forster, Hydrodynamic Fluctuations, Broken Symmetry, and Correlation Functions (Benjamin, Reading, MA, 1975).
[15] This is true if the superconducting order parameter is held fixed while rotating the magnetization. There is a Goldstone mode connected to the invariance under rotating all spins, which is unbroken. Estimates of the stiffness of this Goldstone mode, and its coupling to the spin density, show that the resulting contribution to the dimensionless $\chi_T$ is of $O(\mu^3/k^2)$. Consequently, Eq. (2) will hold for wavenumbers $k^2 \gtrsim \mu^3$, while for $k^2 < \mu^3$, $\chi_T$ will diverge like $1/k^2$ after all. Since wavenumbers on the order of $\mu$ are already hard to observe, this means that for practical purposes $\chi_T$ will appear massive.
[16] M. Aronson, private communication (2003).
[17] This is the asymptotic low-$T$ behavior that results from the order parameter given in Eq. (4) below. For simplicity, we use the asymptotic behavior everywhere below $T_c$. The specific heat discontinuity has been chosen to match the maximum in the experimental data. Theoretical values depend on the symmetry of the order parameter. Typical values in a weak-coupling theory, which are universal, are on the order of 1.2 - 1.4 (i.e., much larger than the height of the observed feature); in strong-coupling theories they are not universal [18].
[18] This choice of the orbital structure of the gap function is entirely arbitrary. Determining theoretically which of the possible structures has the lowest energy would be very difficult. However, since different orbital symmetries lead to different thermodynamic properties, one can distinguish between them experimentally. For instance, an orbital symmetry corresponding to the ABM state in Helium 3 would lead to a specific heat $C(T \to 0) \propto T^3$ rather than $T^2$ [19]. For simplicity we only study the order parameter given in Eq. (4). If experimental evidence should favor a different structure, the theory can be easily adjusted to that.
[19] D. Vollhardt and P. Wölfle, The Superfluid Phases of Helium 3 (Taylor & Francis, 1990).
[20] A ferromagnetic superconductor should have an intrinsic nonzero specific heat coefficient, in analogy to the specific heat in the vortex phase of an ordinary type-II superconductor [21]. Here we neglect this effect, whose size will depend on details of the superconducting phase which are not known.
[21] R. A. Fisher, S. Kim, B. F. Woodfield, N. E. Phillips, L. Taillefer, K. Hasselbach, J. Floquet, A. L. Georgi, and J. L. Smith, Phys. Rev. Lett. 62, 1411 (1989).
[22] K. Hasselbach, L. Taillefer, and J. Floquet, Phys. Rev. Lett. 63, 93 (1989).
[23] S.-K. Ma, Modern Theory of Critical Phenomena (Benjamin, Reading, MA, 1976).
[24] In a diagrammatic language, this is equivalent to a generalized random-phase approximation, as is obvious from the structure of Eq. (4).
[25] D. Belitz, T. R. Kirkpatrick, A. J. Millis, and T. Vojta, Phys. Rev. B 58, 14155 (1998).
[26] E. Brézin and D. J. Wallace, Phys. Rev. B 7, 167 (1973).
[27] A. L. Fetter and P. C. Hohenberg, in Superconductivity, edited by R. D. Parks (Marcel Dekker, New York, 1969), p. 817.