LETTER TO THE EDITOR

Competition between disorder and exchange splitting in superconducting ZrZn$_2$

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Abstract. We propose a simple picture for the occurrence of superconductivity and the pressure dependence of the superconducting critical temperature, $T_{SC}$, in ZrZn$_2$. According to our hypothesis the pairing potential is independent of pressure, but the exchange splitting, $E_{xc}$, leads to a pressure dependence in the (spin dependent) density of states (DOS) at the Fermi level, $D_\sigma(\varepsilon_F)$. Assuming p-wave pairing $T_{SC}$ is dependent on $D_\sigma(\varepsilon_F)$ which ensures that, in the absence of non-magnetic impurities, $T_{SC}$ decreases as pressure is applied until it reaches a minimum in the paramagnetic state. Disorder reduces this minimum to zero, this gives the illusion that the superconductivity disappears at the same pressure as ferromagnetism does.

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The coexistence of ferromagnetism and superconductivity is a problem of long standing and general interest. Thus its recent discovery in UGe$_2$ [1], ZrZn$_2$ [2] and URhGe [3] is attracting considerable attention. In particular its occurrence in ZrZn$_2$ is intriguing because, at ambient pressure, ZrZn$_2$ is a weak ferromagnet ($T_{FM} \approx 28.5$ K) and by the application of pressure it can be tuned through a quantum critical point (QCP) ($P_C \approx 21$ kbar) to become a paramagnetic metal [2]. This revives an old suggestion of Fay and Appel [4]. These authors calculated $T_{SC}$ mediated by paramagnons in a McMillan like formalism and found that there is superconductivity in the (triplet) A$_1$ channel [5] on both sides of the QCP. However, while the broad description of Fay and Appel agrees with the observations the fine details do not. In Fay and Appel’s theory the transition temperature goes to zero at the QCP, $T_{SC}$ then rises to a (local) maxima as the model is tuned away from criticality (experimentally this corresponds to pressure being varied away from $P_C$). $T_{SC}$ then falls again away from the QCP. They also predicted that $T_{SC}$ would be approximately the same magnitude in both the ferromagnetic and paramagnetic sides of the phase diagram (although slightly higher on the paramagnetic side).

When superconductivity was observed in ZrZn$_2$ it was only seen on the ferromagnetic side of the phase diagram and further the maximum in $T_{SC}$ was observed at ambient pressure [2]. The experiments show a monotonic decrease in $T_{SC}$ with

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pressure until about 18 kbar. No data has been published in the pressure range 18 kbar \( P < 22 \) kbar, therefore one cannot ascertain where in this range \( T_{\text{SC}} \) falls to zero. Several groups have attempted to explain this either by revisiting the theory of Fay and Appel and examining specific coupling mechanisms \cite{6} or by considering the problem within a Ginzburg–Landau formalism \cite{7}. Both of these groups predicted that \( T_{\text{SC}} \) goes to zero at \( P_c \). Here we will present a simple alternative to these scenarios for the coexistence of superconductivity and ferromagnetism in \( \text{ZrZn}_2 \). We will show that no variation in the coupling constant with pressure (i.e. proximity to the QCP) is required to explain the experiments due to the natural enhancement of the \( \lambda_1 \) phase transition temperature in the ferromagnetic phase and the extreme sensitivity of the triplet pairing states to scattering from non-magnetic impurities. On the other hand our arguments rely on \( \text{ZrZn}_2 \) being a rare example of a Stoner ferromagnet. Interestingly in our consideration the QCP plays no special role and the pressure at which superconductivity disappears is predicted to be strongly sample dependent.

We wish to consider the problem via the simplest model which has the possibility of illustrating the relevant physical phenomena: triplet superconductivity and ferromagnetism. To this end we study a one band Hubbard model with an effective, attractive, pairwise, nearest neighbour interaction, \( U_{ij\sigma'\sigma} \). We allow ferromagnetism to enter via the Stoner model which appears to be in good agreement with the observed behaviour of the ferromagnetic phase of \( \text{ZrZn}_2 \) \cite{2,8,9,10}. Thus we study the consequences of the following Hamiltonian:

\[
\hat{\mathcal{H}} = -\sum_{ij\sigma} t_{ij} \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} + \frac{1}{2} \sum_{ij\sigma\sigma'} U_{ij\sigma\sigma'} \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma} \hat{c}_{j\sigma'}^\dagger \hat{c}_{j\sigma'} - \sum_{\sigma} \sigma E_{\text{xc}} \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma} \tag{1}
\]

where \( \hat{c}_{i\sigma}^\dagger \) are the usual annihilation (creation) operators for electrons with spin \( \sigma = \pm 1 \) occupying a tight binding orbital centred on the lattice site labelled by \( i \). To render the model tractable we assume that the sites \( i \) form a simple cubic lattice.

For a general triplet pairing state in a field gap equations cannot be derived in the same way as they can for the zero field case \cite{11,12}. However, if we specialise to the case of equal spin pairing (ESP) and neglect the action of the dipolar field on the orbital motion of the electrons a remarkable simplification occurs as shown in reference \cite{12} the gap equations are

\[
\Delta_{\sigma\sigma}(k) = -\sum_{k'} \frac{U_{\sigma\sigma}(k-k') \Delta_{\sigma\sigma}(k')}{2E_{\sigma}(k') \left( 1 - 2f_{E_{k'}} \right)} \tag{2}
\]

where the quasiparticle spectrum is given by

\[
E_{k\sigma} = \sqrt{(\varepsilon_k - \mu - \sigma E_{\text{xc}})^2 + |\Delta_{\sigma\sigma}(k)|^2}. \tag{3}
\]

Equations \( \ref{1} \) and \( \ref{3} \) have several surprising features. Firstly, there is a complete decoupling of the two spin states; even in the presence of Cooper pairing. Secondly, the exchange splitting enters only in the role of a chemical potential, but with opposite signs for the two spin states. It must be stressed that these results are only valid for ESP states. However, the large exchange splitting in a ferromagnet probably precludes opposite spin pairing (OSP) states (that is singlet states, \( S_z = 0 \) triplet states or even the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state, which is only stable for moderate exchange splittings). In short \( \ref{1} \) and \( \ref{3} \) are just the Hartree–Fock–Gorkov
approximations restricted to the ESP sector of the theory. For $U_{ij\sigma\sigma'} = U_{ij}\delta_{\sigma\sigma'}$ no such restriction is required as OSP states are not a possibility.

As $T \to T_{SC}$, $|\Delta_{\sigma\sigma'}(k)| \to 0$ and we find the linearised gap equations:

$$\Delta_{\sigma\sigma}(k) = \sum_{k'} \frac{U_{\sigma\sigma}(k-k')}{2(\varepsilon(k') - \mu - \sigma E_{xc})} \tanh \left( \frac{\varepsilon(k') - \mu - \sigma E_{xc}}{2k_B T} \right) \Delta_{\sigma\sigma}(k') .$$

Before solving these equations numerically we must first choose the hoping integrals, $t_{ij}$, and the coupling constants $U_{ij\sigma\sigma'}$. We fit the hoping integrals with an on site ($t_{ii} = \mu$) and nearest neighbour terms only so as to give the same relative density of states in the region of the Fermi level as is found in \textit{ab initio} band structure calculations \cite{10, 13}. The DOS from our fit is compared with that found in the \textit{ab initio} calculation in figure \textbf{1}. Evidently, our one band model cannot reproduce the complex behaviour of the DOS over the full energy range of the Zr related d-band, nevertheless, within an energy range of 20 meV about the Fermi energy it does do so. Since $U_{ij\sigma\sigma'}$ depends on only one parameter: $U_{ij} = U$ for sites $i$ and $j$ being nearest neighbours $(U_{ij} = 0$ otherwise) it can be determined by reference to the measured $T_{SC}$ for clean ZrZn$_2$. This is hampered by the extreme sensitivity of triplet pairing to scattering from non-magnetic impurities \cite{14, 15} and by the lack of data. Using experimental estimates of the residual resistivity, $\rho$, we find that what little data there is \cite{16} is consistent with a clean superconducting critical temperature, at ambient pressure, $T_{SC}^0(P = 0) \sim 1.2$ K as shown in the inset to figure \textbf{1}.

We solved (4) numerically with $U = 0.88t$ on a k-space integration mesh of $10^9$ points. Such a fine integration mesh is required to accurately reproduce the DOS. Our
Figure 2. The phase diagram of our model. The critical temperature is shown for both $A_1$ and $A_2$ phases over a range of exchange splittings. The hatched area indicates the $A$ phase, which is the ground state when $E_{xc} = 0$.

method (implicitly) requires an accurate calculation of the $D_\sigma(\varepsilon_F)$ as we are varying the exchange splitting and thus we are changing $D_\sigma(\varepsilon_F)$, so any errors in evaluating $D_\sigma(\varepsilon_F)$ will lead to significant errors in our calculation of the variation of $T_{SC}$ with $E_{xc}$.

The results of our numerical calculations are shown in figure 2. The $A_1$ phase displays superconductivity in only the $\uparrow\uparrow$ channel. In the $A_1$ phase $d(k) \sim (k_x + ik_y, i(k_x + ik_y), 0)$, where $d(k)$ is the usual BW vector order parameter for triplet superconductivity [5]. The $A_2$ phase corresponds to superconductivity in both ESP channels but with different amplitudes in the two channels so that $d(k) \sim (k_x + ik_y, \kappa(k_x + ik_y), 0)$ where $0 < \kappa < 1$. The $A$ phase, which is only stable in zero exchange splitting, has the same pairing amplitude for both of the ESP states, corresponding to the order parameter $d(k) \sim (k_x + ik_y, 0, 0)$. Because of the complete separation of the up and down sheets the linearised gap equations (4) can be used to calculate the lower transition temperature, $T_{SC}^{A_2}$. This transition represents the formation of a superconducting state in the minority spin state. Our phase diagram of course assumes that no other phase transitions occur. The existence of the $A_1$ and $A_2$ phases only is consistent with a general symmetry analysis [17] and the Ginzburg–Landau expansion of the free energy [12, 18].

To make contact with experiment we must allow for the strong dependence of the superconducting transition temperature of a p-wave superconductor on non-magnetic impurity scattering [14, 15]. This is done via the Abrikosov–Gorkov formula:

$$\ln \left( \frac{T_{SC\rho}}{T_{SC}} \right) = \psi \left( \frac{1}{2} + \frac{\hbar}{4\pi\tau_{tr}k_BT_{SC}} \right) - \psi \left( \frac{1}{2} \right)$$

(5)

where $\tau_{tr}$ is the quasiparticle lifetime as measured in transport experiments and $\psi(x)$ is the digamma function. Note that we do not need to worry about the Baltensperger–Sarma equation [19, 20], which accounts for the reduction in the critical temperature of a superconductor due to exchange splitting, as this is only valid for OSP states. Thus, the Abrikosov–Gorkov formula can be used to calculate $T_{SC\rho}$ as a function of $\tau_{tr}$, or equivalently $\rho$ via the Drude formula. To make the most of the available experimental
data we used the Abrikosov–Gorkov formula \( E_{xc} \) to investigate the effect of disorder on the above phase diagram (see figure 3).

The final step needed to make a direct comparison with measurements of \( T_{SC} \) as a function of pressure is to note that, experimentally, the Curie temperature is a linear function of pressure \( T_{FM}(P) = T_{FM}(0) (1 - P/P_C) \). The zero temperature magnetisation is also linear in pressure \( M(P,T=0) = M(0,0) (1 - P/P_C) \). For a Stoner ferromagnet, the magnetisation is linearly dependent on the exchange splitting and hence

\[
E_{xc}(P,T=0) = \begin{cases} 
E_{xc}(0,0) \left(1 - \frac{P}{P_C}\right) & P \leq P_C \\
0 & P > P_C.
\end{cases}
\]

We now invoke the fact that \( T_{FM} \gg T_{SC} \) which implies that \( E_{xc}(P,T = T_{SC}) \sim E_{xc}(P,T = 0) \). Thus we can map the results of \( T_{SC}(E_{xc}) \) (shown in figure 6) onto \( T_{SC}(P) \) which we show in figure 4. It can be seen that although quantitative agreement with experiment is not achieved, the general features of experiment are reproduced by several of the curves.

Thus we conclude that we have demonstrated the viability of the following simple picture. Irrespective of the mechanism of pairing and exchange splitting ZrZn\(_2\) is a p-wave superconductor with a low \( T_{SC} \sim 1.2 \) K (at ambient pressure). This superconductivity is not observed in the paramagnetic phase of currently available samples due to disorder. However, the exchange field enhances \( T_{SC} \) of an \( A_1 \) p-wave state and this is the cause of the observed superconductivity in the ferromagnetic phase. Experiment suggests \( ZrZn_2 \) is a rare Stoner ferromagnet for which the exchange splitting is proportional to the magnetic order parameter. Thus when \( P > P_C \) and therefore \( T_{FM} = M = E_{xc} = 0 \) there is no measurable superconductivity. However, improvement in sample quality will lead to a lowering of the residual
resistivity and thus presents the possibility of the observation of superconductivity in the paramagnetic state (as is demonstrated by the curves with $\rho_{tr} < 0.3 \mu\Omega\text{cm}$ in figure 4). This explanation is consistent with and lends microscopic support to the more phenomenological arguments of Walker and Samokhin [7] and Mineev [17].

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