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

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 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.

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). More recent work has focused on superconductivity on the paramagnetic side of a
ferromagnetic QCP [6–8].

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 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_{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 [9] or by considering the problem within a Ginzburg–Landau formalism [10]. Both of these groups predicted that T_{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 $ZrZn_2$. 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 A_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 $ZrZn_2$ 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 $ZrZn_2$ [2, 11–13]. Thus we study the consequences of the following Hamiltonian:

$$\hat{\mathcal{H}} = -\sum_{ij\sigma} t_{ij}\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + \tfrac{1}{2}\sum_{ij\sigma\sigma'} U_{ij\sigma\sigma'}\hat{c}^{\dagger}_{i\sigma}\hat{c}_{i\sigma}\hat{c}^{\dagger}_{j\sigma'}\hat{c}_{j\sigma'} - \sum_{i\sigma} \sigma E_{xc}\hat{c}^{\dagger}_{i\sigma}\hat{c}_{i\sigma} \qquad (1)$$

where $\hat{c}^{(\dagger)}_{i\sigma}$ 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 [14, 15]. However, if we specialize 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 [15, 16], the gap equations are

$$\Delta_{\sigma\sigma}(\boldsymbol{k}) = -\sum_{\boldsymbol{k}'} \frac{U_{\sigma\sigma}(\boldsymbol{k}-\boldsymbol{k}')\Delta_{\sigma\sigma}(\boldsymbol{k}')}{2E_{\sigma}(\boldsymbol{k}')}(1-2f_{E_{\boldsymbol{k}'\sigma}}) \qquad (2)$$

where the quasiparticle spectrum is given by

$$E_{\boldsymbol{k}\sigma} = \sqrt{(\varepsilon_{\boldsymbol{k}} - \mu - \sigma E_{xc})^2 + |\Delta_{\sigma\sigma}(\boldsymbol{k})|^2}. \qquad (3)$$

Equations (2) and (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 (2) and (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 $T \to T_{SC}$, $|\Delta_{\sigma\sigma'}(\boldsymbol{k})| \to 0$ and we find the linearized gap equations:

$$\Delta_{\sigma\sigma}(\boldsymbol{k}) = \sum_{\boldsymbol{k}'} \frac{U_{\sigma\sigma}(\boldsymbol{k}-\boldsymbol{k}')}{2(\varepsilon(\boldsymbol{k}')-\mu-\sigma E_{xc})}\tanh\left(\frac{\varepsilon(\boldsymbol{k}')-\mu-\sigma E_{xc}}{2k_BT}\right)\Delta_{\sigma\sigma}(\boldsymbol{k}'). \qquad (4)$$
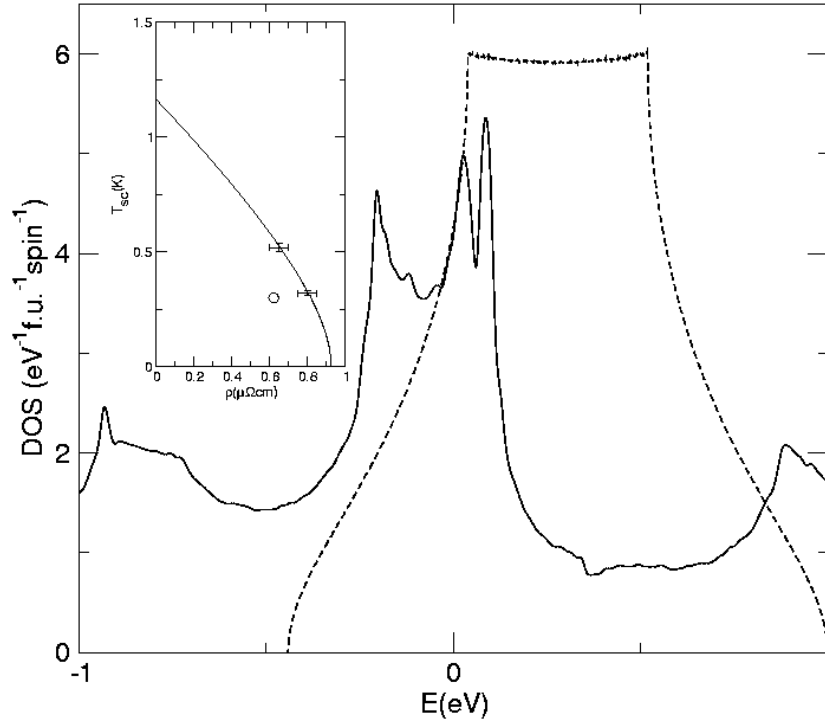
Figure 1. The DOS from the LDA calculations for $ZrZn_2$ (———) by Santi and co-workers [13, 17] and our tight binding fit to the LDA DOS (– – –). Inset: the experimental data [20] is consistent with a fit to $T_{SC}^0(P = 0) \sim 1.2$ K from the Abrikosov–Gorkov formula (5). Also shown (open circle) is a data point taken from [2], we do not fit to this point—see the text for discussion.

Before solving these equations numerically we must first choose the hopping integrals, t_{ij}, and the coupling constants $U_{ij\sigma\sigma'}$. We fit the hopping 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 *ab initio* band structure calculations [13, 17] which have been found to be in excellent agreement with quantum oscillation experiments [13]. The DOS from our fit is compared with that found in the *ab initio* calculation in figure 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. The large cubic Fermi surface of the tight binding model also represents a rough fit to the largest sheet of the LDA Fermi surface (band 29 in the notation of [13]). 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 [18, 19] and by the lack of data. Using experimental estimates of the residual resistivity, ρ, we find that what little data there is [20] 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 1. We do not fit to the data from [2] as in this sample the resistivity does not drop to zero, suggesting an inhomogeneous state. However in more recent experiments the resistivity has been observed to drop to zero [20], indicating that bulk superconductivity is observed.
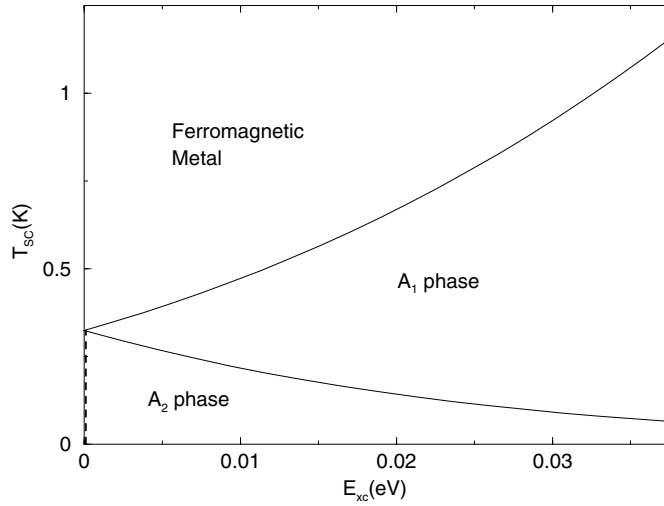
Figure 2. The phase diagram of our model. The critical temperature is shown for both A_1 and A_2 phases over a range of exchange splittings. The hatched area indicates the A phase, which is the ground state when $E_{xc} = 0$.

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 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 $d(k) \sim (k_x + ik_y, i\kappa(k_x + ik_y), 0)$ 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 linearized 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 [21] and the Ginzburg–Landau expansion of the free energy [15, 22].

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 [18, 19]. This is done via the Abrikosov–Gorkov formula:

$$\ln\left(\frac{T_{SC0}}{T_{SC}}\right) = \psi\left(\frac{1}{2} + \frac{\hbar}{4\pi\tau_{tr}k_B T_{SC}}\right) - \psi\left(\frac{1}{2}\right) \tag{5}$$

where τ_{tr} is the quasiparticle lifetime as measured in transport experiments and $\psi(x)$ is the digamma function. The derivation of the Abrikosov–Gorkov formula for a ferromagnet is a straightforward generalization of the derivation in the paramagnetic case if one assumes that τ_{tr} is the same for both spin states due to the complete separation of the spin states in a ferromagnetic superconductor discussed above. Note that we do not need to worry about the Baltensperger–Sarma equation [23, 24], which accounts for the reduction in the critical
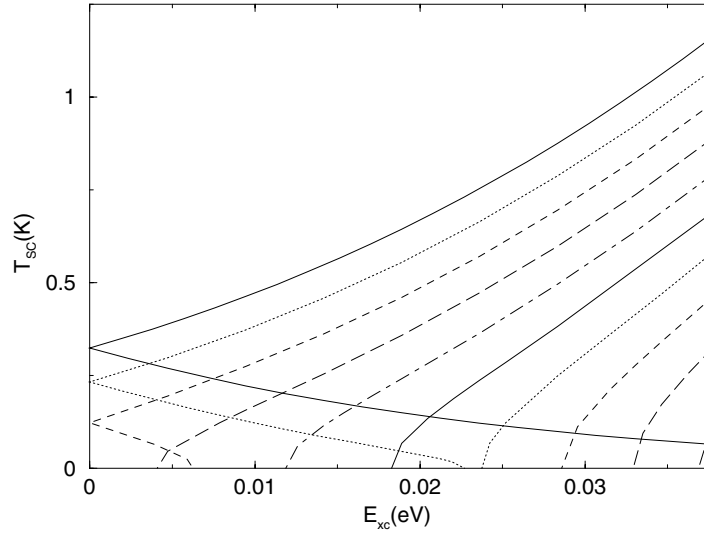
Figure 3. The critical temperature of a ESP p-wave superconductor and the temperature of the
A_1–A_2 transition as a function of exchange splitting in the presence of disorder. The curves
correspond (from the top down) to $\rho_{tr} = 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8$ and $0.9\ \mu\Omega$ cm.

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} as a function of τ_{tr}, or
equivalently ρ via the Drude formula. To make the most of the available experimental data
we used the Abrikosov–Gorkov formula (5) 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 [11, 12]. Namely, $T_{FM}(P) = T_{FM}(0)(1 - P/P_C)$. The zero temperature
magnetization is also linear in pressure [11] and thus proportional to T_{FM} (as is predicted
by the Stoner model), giving $M(P, T = 0) = M(0, 0)(1 - P/P_C)$. For a Stoner ferromagnet,
the magnetization is linearly dependent on the exchange splitting and hence

$$E_{xc}(P, T = 0) = \begin{cases} E_{xc}(0, 0)\left(1 - \dfrac{P}{P_C}\right) & P \leqslant P_C \\ 0 & P > P_C. \end{cases} \tag{6}$$

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 3) 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 [2, 11–13] 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
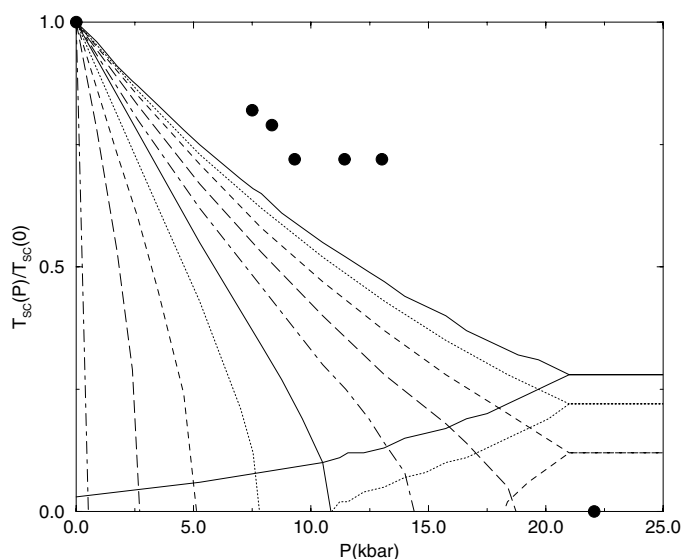superconductivity. However, improvement in sample quality will lead to a lowering of the

Figure 4. The critical temperature of a ESP p-wave superconductor and the temperature of the A_1-A_2 transition as a function of pressure in the presence of disorder. The theoretical curves are scaled so that $T_{SC} = 1$ at ambient pressure. The experimental data, taken from [2], was scaled in the same way. The curves correspond (moving from top right to bottom left) to $\rho_{tr} = 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8$ and $0.9 \, \mu\Omega$ cm.

residual resistivity and thus presents the possibility of the observation of superconductivity in the paramagnetic state (as is demonstrated by the curves with $\rho_{tr} < 0.3 \, \mu\Omega$ cm in figure 4). This explanation is consistent with and lends microscopic support to the more phenomenological arguments of Walker and Samokhin [10] and Mineev [21].

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