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Direct Observation of the Multisheet Fermi Surface in the Strongly Correlated Transition Metal Compound $ZrZn_2$

MAJOR, Zs., *et al.*

Abstract

The existence of flat areas of a Fermi surface (FS), predicted by electronic structure calculations and used in models of both magnetically mediated and phonon-mediated Fulde-Ferrell-Larkin-Ovchinnikov superconducting states, is reported in the paramagnetic phase of the ferromagnetic superconductor $ZrZn_2$ using positron annihilation. The strongly mass-renormalized FS sheet, dominating the Fermi level density of states, is seen for the first time. The delocalization of the magnetization is studied using measured and calculated magnetic Compton profiles.

Reference

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Direct Observation of the Multisheet Fermi Surface in the Strongly Correlated Transition Metal Compound ZrZn_2

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The existence of flat areas of a Fermi surface (FS), predicted by electronic structure calculations and used in models of both magnetically mediated and phonon-mediated Fulde-Ferrell-Larkin-Ovchinnikov superconducting states, is reported in the paramagnetic phase of the ferromagnetic superconductor ZrZn_2 using positron annihilation. The strongly mass-renormalized FS sheet, dominating the Fermi level density of states, is seen for the first time. The delocalization of the magnetization is studied using measured and calculated magnetic Compton profiles.

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The discovery of ferromagnetism coexisting with superconductivity in the compound UGe_2 [1], followed by similar observations in ZrZn_2 [2] and URhGe [3], has reopened the debate on the relationship between magnetism and superconductivity. At its heart is the question of the pairing mechanism for the superconductivity, with some speculation that it could be magnetically rather than phonon mediated. While there have been a number of theoretical models proposing various pairing mechanisms [4–10], experiments have yet to confirm many of the details of the electronic structure on which they are founded.

In this Letter, a combination of experiments and *ab initio* calculations is used to both determine aspects of the topology of the paramagnetic Fermi surface (FS) of ZrZn_2 crucial to the nature of the superconducting state, and investigate the delocalized character of the magnetic moment in the ferromagnetic phase. The implications for the origin of superconductivity in ZrZn_2 are discussed.

Several studies of the electronic structure of ZrZn_2 have been made using *ab initio* techniques based on the local density approximation (LDA) and while some small differences exist among them, all predict that there are four partially filled bands (bands 27 to 30), giving rise to four and eight FS sheets in the paramagnetic and ferromagnetic phases (PM and FM), respectively (see, for example, Ref. [6]). These calculations indicate that about 50% of the density of states (DOS) at the Fermi energy (E_F) is from band 29. The topology of the FS, through the relevance of its nesting properties in the promotion of magnetic excitations which might mediate or stabilize superconductivity [6,7,11–14], is central to any theory.

A recent de Haas–van Alphen (dHvA) study of the FM state has revealed a FS topology in good agreement with

the LDA calculations, albeit with strongly renormalized quasiparticle masses [15]. However, probably due to the combination of large FS orbits and these heavy quasiparticle masses, no conclusive signal attributable to band 29 (which dominates the DOS at E_F) was measured [15]. Furthermore, no direct inferences were possible regarding the flatness (and hence their nesting tendency) of the “pillow” sheets (band 27) [15] (described as “interconnected pancakes” by Singh and Mazin [7]), which is the most likely candidate for the spin-up to spin-down nesting which could be favorable for the development of a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconducting state [2,12], the feasibility of which has been critically investigated [7]. Since the onsets of ferromagnetism and superconductivity in ZrZn_2 appear to be coincident [2], a knowledge of the FS topology is essential for a proper understanding of the development and nature of the superconducting state. Given the extreme sensitivity of the FS topology to the position of the chemical potential, as emphasised in Ref. [7], a precise and complete FS determination is vital.

The paramagnetic FS was obtained from a series of 2-dimensional angular correlation of electron-positron annihilation radiation (2D-ACAR) measurements [16]. The close-packed C15 structure of ZrZn_2 is suited for a 2D-ACAR study since the positron is very uniformly distributed and should thus give information on all the FS sheets. A 2D-ACAR measurement yields a 2D projection (integration over one dimension) of an underlying electron-positron momentum density $\rho(\mathbf{p})$, and the presence of a FS is revealed through the discontinuities in this distribution. Tomographic methods can be used to reconstruct the 3D momentum density from a series of measured projections [17]. While the momentum density is a single-centered distribution having the full

point-symmetry of the crystal lattice in question, the translational invariance of \mathbf{k} space can be restored by “folding” the distribution back into the first Brillouin zone (BZ), giving the 3D occupation density [18]. This technique has recently been used to determine the FS topology and identify nesting features in a wide range of systems [19]. The virtue of the 2D-ACAR technique in such studies is that it reveals *directly* the shape of the FS, important if any propensities for nesting are to be elucidated.

The positron measurements on a single crystal sample of ZrZn_2 [2] were made at ~ 80 K, where the FWHM of the overall momentum resolution of the Bristol 2D-ACAR spectrometer corresponded to $\sim 15\%$ of the BZ. Projections were measured along six different crystallographic directions, namely [100], [110], [210] and 9° , 18° , and 36° from [100] towards [110] in the (001) plane. The individual spectra each contained about 200×10^6 raw counts. A maximum entropy based deconvolution routine [19] was employed to suppress the smearing effect of the finite instrument resolution. From the reconstructed occupation density, the four sheets of the paramagnetic FS have been identified from extrema in the first derivative of that density (Fig. 1), and the corresponding isodensities used to image the FS; these are shown in Fig. 2.

When compared to the calculations of Ref. [7], excellent agreement can be seen in the size and topologies of all four sheets. In particular, the flatness of the pillow sheet (band 27), predicted by the calculation, is clearly observed in experiment. This band, when exchange split

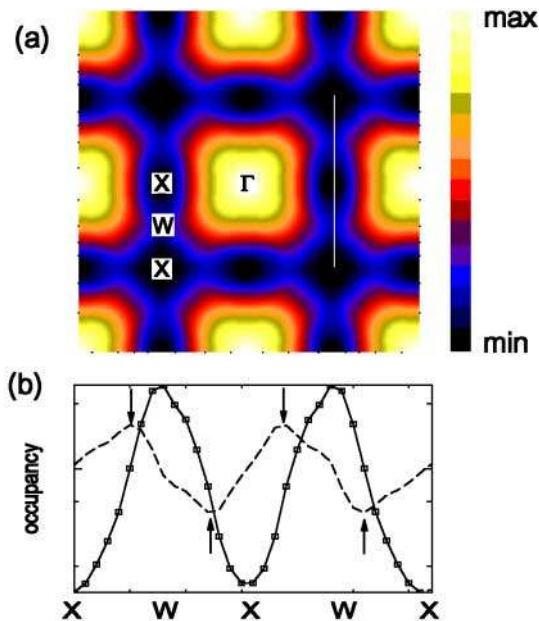


FIG. 1 (color online). (a) The experimental occupancy in the (100) plane of the repeated zone scheme. The colors range from black (low occupancy) to white (high occupancy). (b) Section (solid line) along the white line shown in (a), along with its first derivative (dashed line), used to locate the FS breaks (illustrated for band 27), indicated by the arrows.

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in the FM phase, would be a candidate for supporting the FFLO state [7], although its DOS at E_F is relatively small. Given that no spin or charge-density wave order has been observed in ZrZn_2 , the arguments of Shimahara [20] suggest that these flat surfaces would be ideal for a nesting-enhanced FFLO state. The sensitivity of the FS topology to the position of E_F is apparent when considering the band 29 sheet; that calculated by Singh and Mazin [7] does not have necks at the L points, whereas our experiment shows clear necks. Note that in the calculations of Santi *et al.* in the FM phase [6] the majority sheet of band 29 also possesses a neck, indicating how very small shifts in E_F (or small changes in lattice parameter) could open or close a neck. The flat sections of FS evident in bands 29 and 27 are likely to promote spin fluctuations, which could provide a magnetically mediated pairing mechanism [5,6,13]. In general, the qualitative agreement between experiment and calculation is excellent, indicating the fidelity of the *ab initio* description. However, since an accurate description of the PM electronic structure is essential, a quantitative evaluation was deemed necessary, and carried out by a direct comparison between *ab initio* calculations and the experimental data through a rigid-band-like fitting procedure.

The PM electronic structure was calculated at the experimental lattice constant (13.988 a.u.) using the linearized muffin-tin orbital (LMTO) method within the atomic sphere approximation (ASA), including combined-correction terms [21]. Here the scalar-relativistic (SR) approximation was employed, which does not include any spin-orbit coupling (SOC), in contrast to fully relativistic (FR) calculations which will be discussed later. The details of the calculations are as previously reported [6,15]. In our fitting procedure the four partially occupied bands from this LMTO

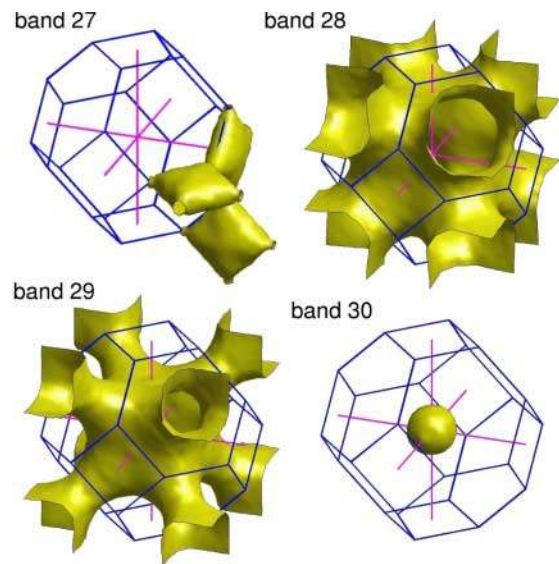


FIG. 2 (color online). The four sheets of FS of paramagnetic ZrZn_2 , obtained from the positron experiment.

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calculation are each allowed to be populated up to a different Fermi energy, and for each shift of E_F the corresponding electron–positron momentum density is determined. Since the FS information is in the anisotropic part of $\rho(\mathbf{p})$, the so-called radial anisotropy is constructed by subtracting from the data [a 2D projection of $\rho(\mathbf{p})$], its radial average [22]. By simultaneously fitting the radial anisotropies of the calculated spectra to those of all six measured projections, a set of rigid-band shifts was obtained, which corresponded to the best agreement between the calculated and measured FS topologies. The FS topology thus produced is, of course, not forced to respect any degeneracies (which are present in the SR calculations, but are lifted when SOC is included). The resulting Fermi level shifts were -6 ± 1 mRy, -7.6 ± 0.5 mRy, $+0.1 \pm 0.5$ mRy, $+7 \pm 5$ mRy for bands 27 to 30, respectively. The errors are largest for the most dispersive bands (in particular, band 30, where the small size and spherical shape of the FS makes its contribution to the radial anisotropy small), but none of the shifts are more than 6% (for 27–30, 4%, 6%, 0.06%, 3%, respectively) of their respective bandwidths. It should be borne in mind that the exchange splitting in the FM phase is ~ 4.5 mRy. Note that the extremal area of band 30 ($0.71 \pm 0.07 \text{ \AA}^{-2}$) obtained from the fit is the mean of the 30 \uparrow and 30 \downarrow areas measured by Yates *et al.* [15]. The (flat) band 29, unseen in the dHvA experiments, but whose topology changes rapidly with E_F , is particularly precisely located. Although we acknowledge the limitation associated with such a rigid-band procedure which does not allow the shape of the bands to change, the shifts of the dominant bands (27–29) are so small that the difference to the real dispersion of the bands can be neglected to a good approximation. The result of these shifts changes the DOS at E_F by less than 0.5%, and thus would not have a large effect on the electron-phonon coupling constant λ , which is of importance in all pairing mechanisms.

To investigate the impact of SOC on the FS, we made self-consistent calculations using the FR Korringa-Kohn-Rostoker (KKR) method [23,24]. For comparison with the LMTO, SR KKR calculations were also performed. The FR spin-polarized calculations predict spin and orbital moments of $0.18\mu_B$ and $-0.0096\mu_B$, respectively. This is in very good agreement with both the moment obtained from the LMTO calculations [6] and the experimental value ($0.17\mu_B$). However, owing to the use of the ASA in both the LMTO and KKR calculations, and given that full-potential calculations predict substantially larger magnetizations [7] this finding should not be over-interpreted [25]. The effect of SOC on the paramagnetic and ferromagnetic FS topologies is shown in Fig. 3. In the FM calculation, the shading indicates the majority/minority spin character. SOC slightly modifies the connectivity of the FS through avoided band crossings, but does not alter the principal features, explaining the good agreement between our SR calculations and the current positron and previous dHvA experiments [15]. Note that

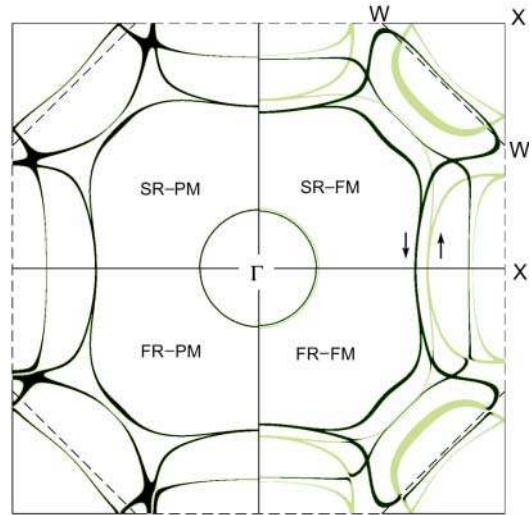


FIG. 3 (color online). FS from fully-relativistic (FR) and scalar-relativistic (SR) KKR calculations in the (001) plane of ZrZn_2 [26]. The paramagnetic (PM) calculations are on the left-hand side, and the ferromagnetic (FM) ones are on the right. Note that the shading in the FM calculations indicates majority (\uparrow) / minority (\downarrow) spin. The magnetization axis is along [001].

the small differences between the SR FS topologies predicted by the LMTO and KKR can be eliminated by very small (less than 1 mRy) shifts in E_F , providing further evidence for the extreme sensitivity of the FS.

In addition to detailed knowledge of the FS topology, the behavior of the magnetic moment is of crucial importance for understanding the connection between superconductivity and magnetism, given the likelihood that the same electrons are responsible for both. Previous neutron experiments [27] have indicated a surprisingly delocalized magnetization distribution in ZrZn_2 . Moreover, *ab initio* calculations predict that the exchange splitting is not uniform over the FS and that it can be very different on each sheet. In order to investigate more closely the composition and delocalization of the spin magnetic moment, a magnetic Compton profile (MCP) was measured. A MCP is a double integration of the spin

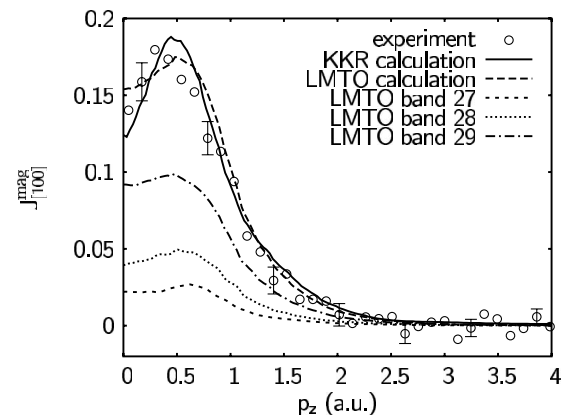


FIG. 4. Magnetic Compton profile along [100].

density in momentum space [28]. The momentum was resolved along the [100] direction at a temperature of 10 K and a magnetic field of 2.5 T (under which conditions the magnetic moment is $\sim 0.2\mu_B$) on beam line BL08W of the SPring-8 synchrotron (Japan). In Fig. 4, the measured profile is plotted alongside those calculated using the (SR) LMTO and the (FR) KKR methods. At this experimental resolution (approximated by a Gaussian of FWHM = 0.45 a.u., and with which the theoretical curves have been convoluted) there are negligible differences between the LMTO and KKR calculations, but from the widths of the profiles it is clear that both calculational frameworks predict a delocalization of the magnetic moment in good agreement with experiment. Since the overall shape of the MCP is characteristic of the localization of each band, such a decomposition (Fig. 4) indicates that the calculations are describing the exchange splitting on each FS sheet rather well.

In conclusion, we have presented the first experimental determination of the FS of paramagnetic ZrZn₂, and shown it to be in good agreement with *ab initio* calculations. In addition to detecting all four FS sheets, including the “heavy” band 29 sheet absent in a previous dHvA study, an innovative analysis method has permitted the calculated and measured FS topologies to be compared to a level unprecedented in a positron annihilation experiment. The inclusion of SOC has been shown to have only a minor effect on the FS topology in both the PM and FM phases. Finally, our measurement of the magnetic Compton profile indicates that the degree of delocalization of the magnetic moment is reasonably well described by the calculations. Our results have implications for the models attempting to explain superconductivity in ZrZn₂. First, a flat FS sheet in band 27 (the pillows) has been revealed which would be an excellent candidate for providing the enhancement [20] to the FFLO superconducting state proposed for ZrZn₂ [2,7]. Second, the band 29 FS, which also has flat surfaces, implies that FS nesting-driven spin fluctuations are likely to be a strong feature, and thus has important consequences for a pairing interaction of magnetic origin [6,13]. Finally, several theories predict the occurrence of superconductivity in the PM phase [9,13]; while this has not yet been observed, the inclusion of a realistic, experimentally proven band structure [29] in such models is likely to shed further light on the validity of these scenarios.

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