# Evolution of Fermi-Liquid Interactions in $Sr_2RuO_4$ under Pressure

D. Forsythe,[1] S. R. Julian,[1] C. Bergemann,[1] E. Pugh,[1] M. J. Steiner,[1] P. L. Alireza,[1] G. J. McMullan,[1] F. Nakamura,[2] R. K.W. Haselwimmer,[1] I. R. Walker,[1] S. S. Saxena,[1] G. G. Lonzarich,[1] A. P. Mackenzie,[3] Z. Q. Mao,[4] and Y. Maeno[4]

[1]*Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 OHE, United Kingdom*
[2]*Graduate School of Advanced Sciences of Matter, Hiroshima University, Higashi-Hiroshima 739-8526, Japan*
[3]*School of Physics and Astronomy, University of St. Andrews, St. Andrews, Fife KY16 9SS, United Kingdom*
[4]*Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan*

(Received 5 June 2002; published 27 September 2002)

We have measured the temperature and field dependence of the resistivity of the unconventional superconductor $Sr_2RuO_4$ at pressures up to 3.3 GPa. Using the Shubnikov–de Haas effect, we find that the Fermi surface sheet believed to be primarily responsible for superconductivity becomes *more* two-dimensional with increasing pressure, a surprising result that is, however, consistent with a recent model of orbital-dependent superconductivity in this system. Many-body enhancements and the superconducting transition temperature all fall gradually with increasing pressure, contrary to previous suggestions of a ferromagnetic quantum critical point at $\sim$3 GPa.

DOI: 10.1103/PhysRevLett.89.166402 PACS numbers: 71.18.+y, 71.20.–b, 71.30.+h

There is now persuasive evidence that the layered perovskite oxide metal $Sr_2RuO_4$ has an exotic superconducting ground state, in which the Cooper pairs form as spin triplets with odd-parity relative angular momentum [1]. Moreover, the normal state from which the superconductivity develops is both simple and well described by Fermi liquid theory [2,3], but the mechanism of superconductivity is not yet settled.

Many theories of the superconductivity of $Sr_2RuO_4$ assume that this material lies close to a magnetic quantum critical point (QCP), and that the superconductivity is mediated by nearly critical magnetic fluctuations [4,5], which should also influence the normal state properties. This provides strong motivation for a new generation of measurements to examine the simultaneous evolution of the superconducting and normal states as a function of pressure, a tool known to be effective in tuning the strength of magnetic correlations without changing the level of disorder in a system [6–8].

There have been two previous pressure studies. Shirakawa *et al.* [9] measured the pressure dependence of the superconducting $T_c$ using resistivity, finding a decrease with pressure at a rate of about 0.3 K/GPa, which in their crystal ($T_c \sim 0.9$ K at ambient pressure) suggests that superconductivity should be suppressed near 3.0 GPa. In the limited pressure range of their experiments, they were unable to resolve any change in the $T^2$ coefficient (commonly denoted $A$ from $\rho = \rho_0 + AT^2$). A second study by Yoshida *et al.* [10] measured the temperature dependence of the resistivity above 4.2 K at pressures up to 8 GPa and made the radical proposal, based on a $T^{4/3}$ power law that emerges in their $\rho(T)$ at high pressure, that $Sr_2RuO_4$ has a quasi-two-dimensional ferromagnetic ground state above $\sim$3.0 GPa, suggesting that pressure is pushing $Sr_2RuO_4$ towards a QCP.

In this Letter, we have carried out a more advanced pressure study of the resistivity of $Sr_2RuO_4$ in which we have simultaneously determined the pressure dependence of the Fermi surface (including detailed examination of the $c$-axis dispersions), many-body mass enhancements, and the superconducting $T_c$. We used Shubnikov–de Haas (SdH) oscillations to monitor the Fermi surface and to obtain the mass enhancements on individual Fermi surfaces, while the $T^2$ coefficient of the resistivity was used to provide an alternative measure of many-body enhancements. These sheet specific measurements are unusual in that they provide a basis for input to theories that focus on the normal state of *orbital-dependent* superconductors, where conventional measurements (such as the $T^2$ coefficient) are less effective since they are restricted to averages over the Fermi surface sheets. In $Sr_2RuO_4$, the orbital-dependent situation is thought to be realized since the different $z$-axis inversion symmetries of the $d_{xz/yz}$ and $d_{xy}$ orbitals, respectively, cause them to form two largely unconnected electronic subsystems.

This model has recently been refined by Zhitomirsky and Rice [11], who argue that part of the $c$-axis dispersion of $Sr_2RuO_4$ reflects the small mixing between the active superconducting band (thought to be the $d_{xy}$ band which forms the $\gamma$ Fermi surface sheet) and the remaining (passive) bands $d_{xz/yz}$ which form the $\alpha$ and $\beta$ sheets. Their approach is important because it provides an appealing explanation for the apparent inconsistency between the gapless order parameter originally expected for $Sr_2RuO_4$ [12] and the body of evidence that has recently emerged in favor of line nodes in the gap function. These authors investigate the orbital-dependent superconductivity that arises when interlayer processes dominate the coupling between the active and passive bands. In this special situation, a nodeless axial $p$-wave order parameter in the active band can induce superconductivity with horizontal lines of nodes in the passive bands: $d'(\mathbf{k}) \propto (k_x + ik_y)\cos(k_z/2)$. Accurate knowledge of the $c$-axis normal state dispersion is therefore linked to understanding the gap symmetry of $Sr_2RuO_4$. It is also relevant to the pairing mechanism, since the degree of two-dimensionality can strongly influence the spin fluctuation spectrum and the superconducting $T_c$.

The single-crystal samples used in this study were grown using the floating-zone method. They were taken from two high purity batches, both with ambient pressure superconducting $T_c$ near 1.5 K. For the quantum oscillation work, the high-pressure environment was achieved using a miniature sapphire-anvil cell with argon as the pressure transmitting medium. Shubnikov–de Haas oscillation measurements were made in an 18 T cryomagnetic facility at temperatures down to 20 mK and at pressures of 0, 2.0, and 3.3 GPa. To determine the pressure dependence of the $T^2$ coefficient of the resistivity, we have taken additional pressure points using a piston-cylinder high-pressure cell with a 50:50 mixture of $n$-pentane/isopentane as the high-pressure medium. A standard four-terminal method was used to measure the resistance, with contacts made using Dupont 6838 Conducting Composition, cured at 450 °C under flowing oxygen (99.999% purity). In our anvil cell measurements, we cannot rule out a small Hall contribution but previous ambient pressure measurements in this material have shown that any quantum oscillatory Hall voltage would be overwhelmed by magnetoresistance oscillations [13].

Figure 1 shows an SdH trace and its corresponding frequency spectrum, measured at 2.0 GPa. The frequency $F$ of each peak in the spectrum is related to an extremal (i.e., maximum or minimum) cross-sectional area $A$ of the Fermi surface measured in planes perpendicular to the applied field, via $F = n\hbar A/2\pi e$ [14], where $\hbar$ is Planck's constant, $n$ is a positive integer, and $e$ is the electron charge. The peaks in Fig. 1 have been labeled according to the Fermi surface from which they arise.

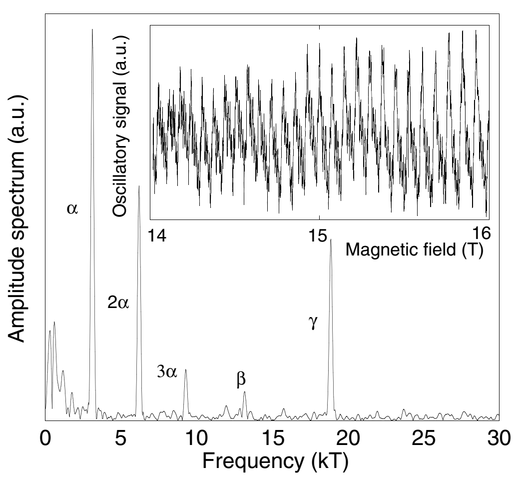


FIG. 1. High resolution Shubnikov–de Haas amplitude spectrum (main) and oscillations (inset) at 1.7 GPa. The $\alpha$ sheet and three additional harmonics can clearly be resolved. The $\beta$ sheet is also visible but is anomalously small in magnitude. The $\gamma$ sheet is clear and makes a much larger contribution than has been seen in samples at ambient pressure.

Up to our highest pressure of 3.3 GPa, the SdH frequencies are only weakly pressure dependent. There is, thus, no sign of a Fermi surface reconstruction, nor of the exchange splitting that would signal the appearance of ferromagnetism at high pressure.

From data such as those shown in Fig. 1, we can extract the pressure dependence of many-body enhancements. Table I shows the change in quasiparticle masses for each sheet of the Fermi surface between ambient pressure and 3.3 GPa, extracted from the temperature dependence of Shubnikov–de Haas oscillations. The highest resolution is for the $\alpha$ sheet, because the oscillations from that surface are the strongest (see Fig. 1) and, having a lighter mass, are observed over the broadest temperature range. For this surface, $m^*$ is found to fall by around 20% between 0 and 3.3 GPa. Because for the $\beta$ sheet the signals die out rapidly with increasing pressure the errors are large, but the rate of decrease is consistent with that found on $\alpha$. For the $\gamma$ sheet the rate of decrease appears to be somewhat smaller. We have carried out local density approximation band-structure calculations for $Sr_2RuO_4$ using the measured pressure dependence of the lattice parameters [15] and find that within the resolution of the calculation the effective mass does not change, so the observed evolution of $m^*$ is (as expected) due to many-body effects, not to a change in the underlying band structure.

Figure 2 shows the suppression of $T_c$ as a function of pressure for our sample compared with the lower-quality sample of Shirakawa *et al.* [9]. The upper dashed line in Fig. 2 is a best fit to the points from our clamp-cell measurements. The lower dashed line is a rescaling of the upper line, based on the Abrikosov-Gorkov model for suppression of superconductivity by pair-breaking impurities ([16] and references therein), in which we have used only one parameter which is related to the strength of impurity scattering, showing that such scattering is pressure independent and again ruling out any exotic evolution of the Fermi liquid state with pressure. The inset in Fig. 2 shows that the $T^2$ coefficient of resistivity falls roughly linearly with pressure by ~45% between 0 and 3.0 GPa, with consistency being obtained between four different samples, in our two different kinds of pressure cells. These results paint a consistent picture of a fall in the Fermi liquid quasiparticle-quasiparticle interaction strength accompanying a fall in $T_c$ as the

TABLE I. The pressure dependence of the quasiparticle effective masses.

| | $\alpha$ | $\beta$ | $\gamma$ |
|---|---|---|---|
| $m^*$ suppression ($m_e$/GPa) | $0.18 \pm 0.01$ | $0.6 \pm 0.4$ | $0.4 \pm 0.3$ |
| % change over 3.3 GPa | $18 \pm 3$ | $30 \pm 20$ | $9 \pm 7$ |

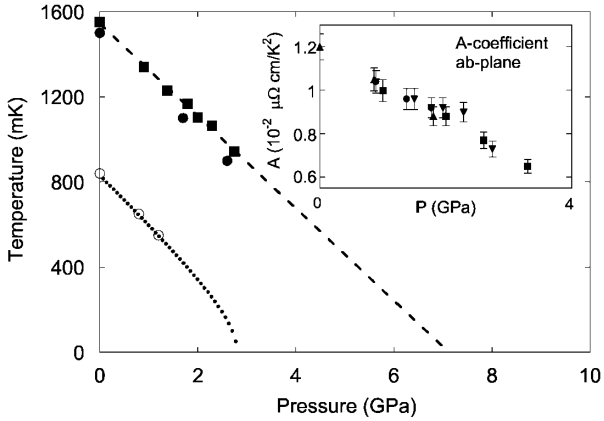


FIG. 2. The superconducting transition temperature and $T^2$ coefficient of resistivity are suppressed under pressure (main and inset, respectively). Filled symbols represent data points from this work (different shapes indicating independent runs) and open circles those from [9]. The upper dashed line in the main plot is a linear fit to the piston-cylinder cell measurements (boxes). The lower dashed line is a rescaling of the upper line, based on the Abrikosov-Gorkov model for the suppression of superconductivity by impurities.

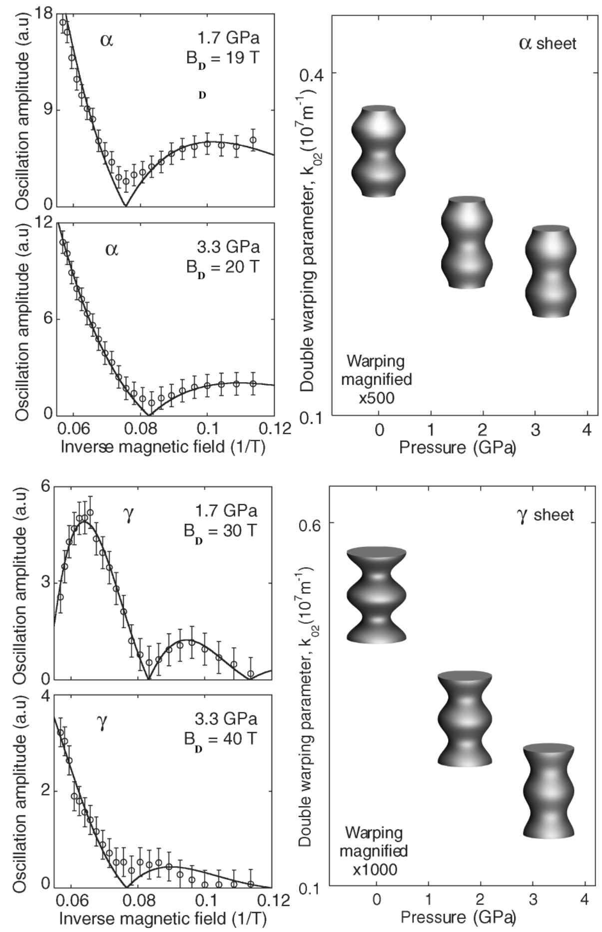


FIG. 3. The $c$-axis coupling in the $\gamma$ sheet, which shows up as a double Fermi surface warping in both the $\gamma$ and $\alpha$ sheets, is seen to reduce under pressure. In the above figure, the theoretical fits used to extract the size of this warping (the $k_{02}$ warping parameter) and the Dingle field, $B_D$, are shown on the left for two pressures (upper diagrams for $\alpha$, lower for $\gamma$), with the resulting pressure dependent warping parameter shown for each sheet on the right.

pressure is increased. In the simplest model of an isotropic interaction in the Landau Fermi liquid theory, the $T^2$ coefficient of resistivity varies as the square of the mass-enhancement $\lambda$, where $m^* = (1 + \lambda)m_{\mathrm{band}}$. Indeed, this seems to hold rather well for the pressure dependences in Fig. 2 and Table I. We note that a suppression of electron-phonon interactions would produce a fall in $m^*$ but no change in the $T^2$ coefficient. Our results thus show that the primary effect of pressure is to suppress electron-electron interactions. Together with the fall in $T_c$, this supports the hypothesis that electron-electron interactions drive the superconductivity.

Combining this information with the absence of non-Fermi liquidlike behavior at any of the pressures (such as a breakdown of the $T^2$ resistivity) and the fact that $T_c$ shows no sign of being suppressed to zero at a unique pressure near 3.0 GPa in our higher purity samples, we can say with certainty that the 2d-ferromagnetic quantum critical point near 3.0 GPa, postulated by Yoshida *et al.* [10], does not exist.

The next interesting issue raised by our measurements concerns the pressure dependence of the $c$-axis dispersion, particularly that of the $\gamma$ sheet, which we determined using nodes in the SdH oscillations (see Fig. 3). The strength of the $c$-axis hopping in a quasi-two-dimensional metal such as $Sr_2RuO_4$ is directly manifested in departures from perfect uniformity of the cross section of the Fermi tubes. These so-called "warpings" can be expressed as an expansion in cylindrical harmonics with coefficients $k_{\mu\nu}$ [3]. As we discuss below, direct interplane overlap leads to "single" warpings of the form $k_{\mu 1}$ and indirect interband-interplane interactions can lead to "double" warpings $k_{\mu 2}$. The warpings have been mapped in great detail in $Sr_2RuO_4$ at ambient pressure by angle-resolved measurements [3], which show that the $\alpha$ and $\beta$ surfaces carry the bulk of the $c$-axis current via mostly single warpings, while $\gamma$ is the most two-dimensional with a small but dominant $k_{02}$ double warping.

Our measurements, which have the field along the $c$ axis, are dominated by contributions from the $k_{02}$ warping component for both the $\alpha$ and $\gamma$ sheets [3]. The size of this component, and thus the degree of two-dimensionality of the $\gamma$ sheet, can be accurately determined from the difference in the maximum and minimum Fermi surface cross-sectional areas, $A_{\max}$ and $A_{\min}$, as a function of $k_z$, the wave vector along the low-conductivity $c$ axis. For both sheets, the nodes that appear in the field sweeps shown in Fig. 3 indicate beating between the signals that arise from these two extremal areas. The nodes move farther apart with increasing pressure, which means that $A_{\max}$ and $A_{\min}$ are getting closer together or, in other words, double warpings are being suppressed in both sheets and the $\gamma$ surface is consequently becoming more two-dimensional at high pressure.

Indications consistent with this conclusion are apparent in the pressure dependence of the SdH signal intensity. The intensity of the $\beta$ signal falls monotonically: it is much weaker at 2.0 GPa than at ambient pressure, and by 3.3 GPa it has disappeared completely. The $\gamma$ signal, in contrast, is much *stronger* at 2.0 GPa than at ambient pressure, although it weakens somewhat at 3.3 GPa. We would normally expect the intensity of quantum oscillation signals to fall with increasing pressure due to pressure inhomogeneity which causes the oscillations from different parts of the crystal to be out of phase and thus to interfere destructively. An increase in two-dimensionality (decreased curvature) is the simplest explanation for an increase in the relative intensity of this sheet under pressure.

A pressure induced increase in the two-dimensionality of the $\gamma$ sheet is counterintuitive because hybridization between orbitals on adjacent planes must increase as the $c$ axis contracts under pressure, and this should equate to increased $c$-axis hopping. Zhitomirksy and Rice [11] have pointed out, however, that the $c$-axis hopping of electrons on the $\gamma$ surface is unusual because Ru 4-$d_{xy}$ orbitals on neighboring planes cannot hybridize due to details of the crystal structure. $c$-axis transport on the $\gamma$ surface is thus a second-order process in which a hop takes place from a state $|\vec{k}\rangle$ on one layer to a state $|\vec{k}\rangle$ *two* layers away, via the $d_{xz,yz}$ orbitals on the intervening layer. The matrix element for this second-order process is $[t''_\perp]^2/[\epsilon_\gamma(\vec{k}) - \epsilon_{\alpha,\beta}(\vec{k})]$, where $t''_\perp$ is the overlap integral between $d_{xy}$ states and $d_{xz,yz}$ states on neighboring planes, and $\epsilon_\gamma(\vec{k})$ and $\epsilon_{\alpha,\beta}(\vec{k})$ are the in-plane dispersion relations for the $\gamma$ band and $(\alpha, \beta)$ bands, respectively. The consequences of this unusual $c$-axis hopping are that (a) the warping on the $\gamma$ sheet is twice as rapid (a double warping) as the dominant warpings on the other sheets, because the hopping path effectively connects *next*-nearest $RuO_2$ planes, and (b) the warping will be largest when $\epsilon_\gamma(\vec{k})$ and $\epsilon_{\alpha,\beta}(\vec{k})$ are most nearly degenerate, i.e., on the zone diagonals.

The first prediction is in agreement with the results of Bergemann *et al.* [3] and is backed up by our findings for the $\alpha$ sheet (Fig. 3), which shows a complementary double warping component, as expected, associated with the same interband-interplane coupling that causes the double warping on $\gamma$. In particular, both components show the same unusual decrease under pressure.

A decrease of this interlayer effect under pressure now appears plausible: the bandwidths and Fermi velocities are expected to increase as the atoms move closer together—and as the band renormalization effects become weaker (see above). Amplified by differential effects, especially along the zone diagonals where $\beta$ and $\gamma$ are close, $|\epsilon_\gamma(k) - \epsilon_{\alpha,\beta}(\vec{k})|$ might then indeed grow more rapidly than $t''^2_\perp$, leading to a *reduction* in the warping.

This, to our understanding, is the only simple explanation for the increased two-dimensionality of the $\gamma$ sheet at high pressure, and as such our results lend strong support to this feature of the model of Zhitomirsky and Rice.

In summary, we have measured Shubnikov–de Haas oscillations in $Sr_2RuO_4$ up to 3.3 GPa. From the slow suppression of superconductivity and the absence of diverging mass renormalizations, we can rule out the postulated existence of a quantum critical point near 3 GPa. In fact, we observe a consistent *suppression* in many-body interactions both as extracted from $m^*$ in quantum oscillation measurements for each sheet and from the $T^2$ coefficient of the resistivity, implying reduced magnetic correlations with increasing pressure. This suppression is accompanied by an *increase* in the two-dimensionality of the $\gamma$ sheet, as is evidenced both in the beating patterns of the quantum oscillations and in the anomalously large relative oscillation amplitude observed for this sheet under pressure. This unusual behavior is consistent with the model of orbital-dependent superconductivity of Zhitomirsky and Rice. Taken together, our results place strong constraints on future microscopic theories of the superconductivity of $Sr_2RuO_4$.

This work was supported by the U.K. EPSRC. In addition, E. P. and A. P. M. acknowledge financial support from the Royal Society of the U.K.

[1] Y. Maeno *et al.*, Phys. Today **54**, No. 1, 42 (2001).
[2] A. P. Mackenzie *et al.*, Phys. Rev. Lett. **76**, 3786 (1996).
[3] C. Bergemann *et al.*, Phys. Rev. Lett. **84**, 2662 (2000).
[4] P. Monthoux and G. G. Lonzarich, Phys. Rev. B **63**, 054529 (2001).
[5] I. I. Mazin and D. J. Singh, Phys. Rev. Lett. **82**, 4324 (1999).
[6] S. S. Saxena *et al.*, Nature (London) **406**, 587 (2000).
[7] N. D. Mathur *et al.*, Nature (London) **394**, 39 (1998).
[8] C. Pfleiderer *et al.*, Phys. Rev. B **55**, 8330 (1997).
[9] N. Shirakawa *et al.*, Phys. Rev. B **56**, 7890 (1997).
[10] K. Yoshida *et al.*, Phys. Rev. B **58**, 15 062 (1998).
[11] M. E. Zhitomirsky and T. M. Rice, Phys. Rev. Lett. **87**, 057001 (2001).
[12] T. M. Rice and M. Sigrist, J. Phys. Condens. Matter **7**, 643 (1995).
[13] A. P. Mackenzie *et al.* (unpublished).
[14] D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, 1976).
[15] O. Chmaissem *et al.*, Phys. Rev. B **57**, 5067 (1998).
[16] A. P. Mackenzie *et al.*, Phys. Rev. Lett. **80**, 161 (1998).