Cyclotron Resonance in the Layered Perovskite Superconductor Sr_2RuO_4

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We have observed cyclotron resonance in the layered perovskite superconductor Sr_2RuO_4. We obtain cyclotron masses for the α, β and γ Fermi surfaces of $(4.33\pm0.05)m_e$, $(5.81\pm0.05)m_e$ and $(9.71\pm0.2)m_e$ respectively. The appreciable differences between these results and those obtained from de Haas- van Alphen measurements are attributable to strong electron-electron interactions in this system. Our findings appear to be consistent with predictions for an interacting Fermi liquid; indeed, semi-quantitative agreement is obtained for the electron pockets β and γ.

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The perovskite superconductor Sr_2RuO_4 is currently the subject of intense research activity [1]. Initial interest was driven by the structural similarities between this compound and the high-T_c cuprates [2]. However, a clear picture has since emerged in which it apparently belongs to an entirely different class of superconductor. In particular, Sr_2RuO_4 shares many properties with liquid ^{3}He. For example: the normal state conforms to the behavior expected in a strongly correlated Fermi liquid (FL) [3,4]; the highest reported T_c is still relatively low (< 1.5 K); T_c is extremely sensitive to non-magnetic impurities [5]; and there is mounting evidence supporting the view that the superconductivity is induced by ferromagnetic fluctuations specific to quasi-two-dimensionality, resulting in spin-triplet pairing [6,7].

One of the many attractive features of the title compound has been the availability of high quality single crystals. This has enabled experimental investigations which have not been possible in the high-T_c counterparts, including de Haas - van Alphen (dHvA) and Shubnikov - de Haas (SdH) measurements [3,8]. Consequently, an extensive body of experimental data has given rise to an increasingly coherent picture of the normal state electronic structure [8] which is in broad agreement with the calculated band structure obtained by Local Density Approximations (LDA) [9]. However, as with many strongly correlated electron systems, there is a considerable discrepancy between the calculated and measured density of states at the Fermi energy (E_F); the band masses (m_b) for the three Fermi surface (FS) pockets (two electron and one hole), estimated from LDA calculations, are considerably smaller (by factors of between 3 and 5) than the thermodynamic masses (m^*) determined from dHvA measurements [3]. These differences have been attributed to the FL corrections expected in an interacting Fermi system.

The aim of the present study is to measure the cyclotron masses (m_c) in Sr_2RuO_4 using a long wavelength probe ($Q\sim0$) which couples to the center-of-mass motion of the system. For a translationally invariant isotropic FL, the band mass, cyclotron mass and thermodynamic mass represent different physical quantities [10,11]. The thermodynamic mass includes enhancements due to the fact that, as quasiparticles move through a medium, they experience a drag force resulting from the displacement of other quasiparticles. Thus, in strongly interacting Fermi systems, m^* includes corrections not ordinarily included in band calculations. A cyclotron resonance (CR) experiment is insensitive to these FL effects due to the absence of quasiparticle drag in the center-of-mass frame. Therefore, a comparison between the CR mass m_c, and the thermodynamic mass m^* (as measured *e.g.* in a dHvA experiment), offers the unique possibility of gauging the magnitude of these FL corrections. We wish to point out, however, that the CR mass (sometimes referred to as the dynamical mass) *is* sensitive to other many-body effects, *e.g.* from phonon and Coulomb interactions (distinct from FL effects, see ref. [10]). Consequently, one expects some enhancement of the CR masses (m_c) relative to the band masses (m_b).

dHvA experiments on Sr_2RuO_4 have confirmed the existence of three quasi-two-dimensional FSs whose cross sections are weakly modulated ($< 1.5\%$ variation in k_F) along the c-direction [3,8,12]. Application of a magnetic field causes carriers to orbit these roughly cylindrical FSs in a plane perpendicular to the field. The dominant result is cyclotron motion within the conducting layers, even when the field is tilted well away from the c-axis. It is this cyclotron motion that one usually couples to in a CR experiment. A more subtle effect concerns the influence of a magnetic field on the carrier velocities parallel to the

cylinder axes. Depending on the field orientation, and on the symmetry of the warping, the c-axis velocities (v_c) will also oscillate (for a detailed discussion, see ref [13]); it is precisely these v_c modulations which are responsible for the Angle-dependent Magnetoresistance Oscillations (AMRO) observed recently in Sr_2RuO_4 [14]. In principle, it should also be possible to couple directly to these periodic v_c modulations in an AC measurement, as has recently been demonstrated for several low-dimensional organic conductors [15,16]. Cyclotron-like resonances observed by this method offer a powerful means (over an above AMRO) of determining the precise FS topologies of low-dimensional conductors [13,17]. With this in mind, we opted for an experimental configuration (see below) which is sensitive to both the in-plane and inter-layer conductivities, σ_{ab} and σ_c respectively.

Because of the large effective masses in Sr_2RuO_4, it was necessary to conduct experiments at the lowest frequencies allowed by the constraint $\omega\tau > 1$, and to work at high magnetic fields. The high quality (long τ) of the Sr_2RuO_4 single crystal used in this study, which was grown by a floating zone method [4] and has a T_c of 1.44 K (mid-point), enabled measurements in the mm-wave spectral range. Fields of up to 33 tesla were provided by the resistive magnets at the National High Magnetic Field Laboratory in Florida.

One of the benefits of working at GHz frequencies is the possibility of utilizing an extremely sensitive cavity perturbation technique [18,19]. Two different cylindrical copper cavities ($\phi \sim$ 10mm, height $\sim$ 10mm) were used in transmission, providing four TE01n (n = 1, 2 and 3) modes in the desired frequency range. Loaded cavity Q-factors ranged from 5×10^3 to 2×10^4, depending on the mode. A single Sr_2RuO_4 crystal (dimensions $\approx 2.5 \times 1 \times < 0.2 mm^3$) was placed close to the bottom of the cavity, half way between its axis and its perimeter, thereby ensuring optimal coupling to the radial AC magnetic fields ($\tilde{\mathbf{H}}_1$) for a given TE01n mode. In this configuration ($\tilde{\mathbf{H}}_1 // ab$-plane), the microwave fields excite both in-plane and inter-layer currents (see inset to Fig. 1 and ref. [18]). The applied DC magnetic field ($\mathbf{B}_o$) was directed along the cavity axis and, therefore, parallel to the sample c-axis, *i.e.* $\tilde{\mathbf{H}}_1 \perp \mathbf{B}_o$. As a spectrometer, we used a Millimeter-wave Vector Network Analyzer (MVNA) [18]. Finally, the cavity containing the sample could accurately and controllably be maintained at any temperature between 1.4 K and 30 K.

Fig. 1a shows changes in absorption within the cavity as a function of magnetic field for several temperatures in the range 1.4 to 6 K. The cavity was excited at 76.4 GHz, which corresponds to its TE013 mode. The data were obtained after subtracting a background cavity response, and have been offset for the sake of clarity. It is apparent that on cooling below $\sim$5 K, a series of absorption peaks develop (indicated by arrows) and grow stronger. To within the confidence of the Lorentzian fits to the data, the peak positions appear to be independent of temperature. The number of Lorentzians (5 at T=1.4K), and the initial estimates of their peak positions, were chosen after inspecting data obtained at four different frequencies (see Fig. 3) − otherwise, all fitting parameters were free running. Fig. 1b shows an almost identical data set obtained at the lower frequency of 64.0 GHz, which corresponds to the TE013 mode of the second cavity. It should be noted that all of the absorption peaks have shifted to lower magnetic field.

Similar data were obtained at 44.5 GHz and 58.5 GHz, corresponding to the TE011 and TE012 modes of the first cavity respectively. However, the absorption peaks were less clearly resolved from each other at these lower frequencies due to the lower $\omega\tau$ product. In the inset to Fig. 2, we show a magnified portion of the high field 44.5 GHz data (highest Q mode); SdH oscillations are clearly visible. A fourier transform gives rise to a single peak (main part of Fig. 2) at a frequency of 2975 T, in good agreement with the α-frequency reported previously [3]. Thus, we can be extremely confident that we are well coupled to the sample within the cavity. Weaker SdH oscillations (due to lower Q-values) were discernible in all but the highest frequency data. The absence of β and γ frequencies in the SdH spectra is due to the relatively high temperature (1.4 K) of these measurements.

Fig. 3 shows a compilation of the absorption peak-field positions plotted against frequency for the four TE01n cavity modes used in this study. The exact peak-field positions were obtained from fits to 1.4 K data. All of the points fall nicely on one of several straight lines which pass through the origin, as expected for cyclotron-like resonances, *i.e.* the resonance fields ($B_{res} = m_c\omega/e$) scale linearly with frequency.

In order to interpret the data, it is first necessary to determine the mechanism responsible for the apparent resonant dissipation within the cavity. Due to the highly anisotropic conductivity in Sr_2RuO_4, one can expect the inter-layer currents to penetrate considerably further into the sample than the in-plane currents, as illustrated in the inset to Fig. 1. At 1.5 K and 50 GHz, we estimate a penetration depth for ab-plane currents of $\delta_{ab} \approx 0.2$ μm; the corresponding inter-layer penetration depth (δ_c) is expected to be 30 to 40 ($\sim [\sigma_{ab}/\sigma_c]^{1/2}$) times greater. In this so-called "skin depth" regime, when currents only flow close to the sample surface, dissipation per unit area is governed by the surface resistance R_s ($\propto \mathrm{Re}\{\sqrt{1/\tilde{\sigma}}\}$) [19]. Assuming a quasi-static approximation, it is easy to see that for screening of the AC fields ($\tilde{\mathbf{H}}_1$) to occur within the bulk of the sample, in-plane currents must flow predominantly over the large flat surfaces, while inter-layer currents will flow at the sample edges (see inset to Fig. 1). When we take into account a geometrical factor ($\sim$ 20 for the sample used in this study), corresponding to a ratio of the areas of the sample faces to its edges,

we find that σ_{ab} and σ_c contribute more-or-less equally to dissipation within the cavity. This contrasts the situation in layered organic conductors, where the inter-layer conductivity tends to dominate such a measurement.

Next, we consider the symmetries of the three FSs in Sr_2RuO_4, and the possible resonance modes in σ_{ab} and σ_c resulting from the application of a DC field $\mathbf{B}_o//c$. Three fundamental cyclotron modes should dominate σ_{ab}, with cyclotron frequencies given by $\omega_{ci} = e\mathbf{B}_o/m_{ci}$, where the subscript i refers to the band, *i.e.* α, β or γ [3]. Depending on the cross sectional shape of each FS section, σ_{ab} may additionally contain odd ($N = 3, 5, etc.$) harmonics of the three fundamental CR frequencies [17]; these harmonics would occur at $1/N$ of the field of the fundamental. Resonances in σ_c are governed by the FS warpings, which have recently been measured by Bergemann *et al.* [12]. The strongest contributions to these warpings have cylindrical symmetry and do not, therefore, affect motion in the c direction. Nevertheless, weaker two-fold (α) and four-fold (α, β and γ) symmetric components do exist [12]. Consequently, σ_c, may display weak resonances at even N harmonics of the fundamental α CR frequency, and $N = 4p$ (p =integer) harmonics of the fundamental β and γ CR frequencies. The fundamental frequencies themselves should not be observable in σ_c [12,13].

We can now attempt to account for all of the observed resonances in terms of the three bands (α, β and γ) in Sr_2RuO_4. The inset to Fig. 3 shows an enlarged view of the low field (high $1/\mathbf{B}$) portion of the $f = 76.4$ GHz data plotted versus ω/ω_c, where $\omega = 2\pi f$ and ω_c is the cyclotron frequency ($e\mathbf{B}/m_c$). The resonances labeled $N = 1$ and $N = 2$ correspond respectively to the up ($\triangle$) and down (∇) triangle data points in the main part of the figure. It is apparent that the $N = 1$ peak is actually the first in a harmonic series with resonances visible up to $N = 4$. Thus, we attribute all of these peaks to a single FS. Further confirmation of this can be found from fits to the data in the main part of Fig. 3 where the slopes obtained for the up and down triangle data points differ by a factor of exactly 2 (± 0.05).

The remaining resonances are not related harmonically, either to each other, or to the data in the inset to Fig. 3. Thus, we conclude that the CR response is attributable to three independent carrier types − in agreement with both the theoretically [9] and experimentally determined FS [8]. The most likely scenario is that the three main resonance peaks (above 8 tesla in Fig. 1) correspond to the three fundamental CR modes associated with σ_{ab}. We assign the heaviest CR mass $m_{c\gamma} = (9.71 \pm 0.2)m_e$ to the γ-FS which, to our knowledge, is the largest electronic effective mass thus far detected by magnetic resonance. The CR mass for the β-FS is $m_{c\beta} = (5.81 \pm 0.05)m_e$, while we attribute the harmonic series of resonances to the α-FS with a CR mass $m_{c\alpha} = (4.33 \pm 0.05)m_e$. These values were deduced from the slopes of the lines through the data in Fig. 3, thereby avoiding any possible (systematic) errors associated with the Lorentzian fits. Interpretation of the main absorption peaks as σ_c resonances would imply CR masses four times greater than those deduced above, thus, effectively ruling out this scenario. At this stage, it is not possible to ascertain whether the α harmonics correspond to σ_{ab} or σ_c resonances; angle dependent studies should be able to resolve this issue [13,17]. The strong harmonic content of the α CR is not unexpected given its approximately diamond shaped cross section [9].

A comparison between the cyclotron masses deduced in this study and the thermodynamic masses ($m^*_\alpha = 3.4m_e$, $m^*_\beta = 7.5m_e$ and $m^*_\gamma = 14.6m_e$) measured by Mackenzie *et al.* [8], reveal clear enhancements of m^* over m_c for the electron-like FSs (β and γ). This does not appear to be the case for the hole-like (α-) FS. However, recent theory by Kanki *et al.* [11] has shown that if the translational invariance of a FL is broken (*e.g.* due to the lattice), m_c may exceed m^* under some circumstances.

According to FL theory, the ratio between m^* and the so-called dynamical mass which, in this case, we are assuming corresponds to the cyclotron mass m_c, is given by $m^*/m_c = 1 + F^s_1/3$, where F^s_1 is the dimensionless $l = 1$ FL parameter [10]. In turn, F^s_1 should be proportional to the thermodynamic density of states at E_F which, for a two-dimensional system, is proportional to the thermodynamic effective mass at E_F, *i.e.* $F^s_1 \propto m^*$. In fact, the T-linear coefficient of the specific heat is in excellent agreement with that estimated from m^* on the assumption of the FS two-dimensionality [8]. Consequently, one expects the mass enhancements $[(m^*/m_c)-1]$ to scale with the thermodynamic masses m^*. Interestingly, a comparison between our results and the dHvA masses reported by Mackenzie *et al.* [8], reveal that the mass enhancements for the β and γ FSs scale most closely with the cyclotron masses $m_{c\beta}$ and $m_{c\gamma}$, *i.e.* $[(m^*_\gamma/m_{c\gamma})-1]/[(m^*_\beta/m_{c\beta})-1] = 1.73$ and $m_{c\gamma}/m_{c\beta} = 1.67$. Nevertheless, the thermodynamic mass ratio ($m^*_\gamma/m^*_\beta = 1.95$) is not too far off from 1.73 either. Indeed, if one uses the β dHvA mass ($7.2m_e$) from ref. [21] and the γ dHvA mass from ref. [8], the mass enhancement ratio and the thermodynamic mass ratio both come out close to 2. All said and done, the very fact that the β and γ mass enhancements seem to scale with the effective masses lends strong support to the assertion that Sr_2RuO_4 is a correlated Fermi liquid.

A word of caution is appropriate at this point. The underlying theory used in the above analysis was developed for a single-band isotropic FL. In Sr_2RuO_4, one might expect a strong coupling between the various cyclotron modes corresponding to each electron or hole band. In turn, these couplings might be expected to have a pronounced effect on the measured cyclotron frequencies, over and above the effects expected for a simple FL. With this in mind, it is reasonable to expect the dominant

electronic modes (β and γ) to have more of an influence on the hole mode (α) [22], as opposed to the other way round, which might explain why the α-data do not follow the same trends seen for the β and γ FSs.

Finally, we consider the temperature dependence of the resonances, which attenuate dramatically above 3 K. This is not expected for a single non-interacting parabolic band, unless the quasiparticle lifetime τ depends strongly on temperature. One explanation, therefore, is that the T^2 dependence of $1/\tau$ is responsible for this attenuation [4]. However, we cannot rule out the possibility that above $\sim$3 K, when k_BT exceeds $\hbar\omega_c$, the CR lines broaden due to non-parabolicity. Interactions would then further attenuate the resonances due to the broadened distribution of cyclotron frequencies.

In summary, we have measured the cyclotron masses corresponding to the α, β and γ FSs in Sr_2RuO_4. Comparisons between these values and those deduced from dHvA studies reveal considerable enhancements of the thermodynamic effective masses which we attribute to electron-electron interactions. Qualitatively, our findings are in good agreement with FL theory.

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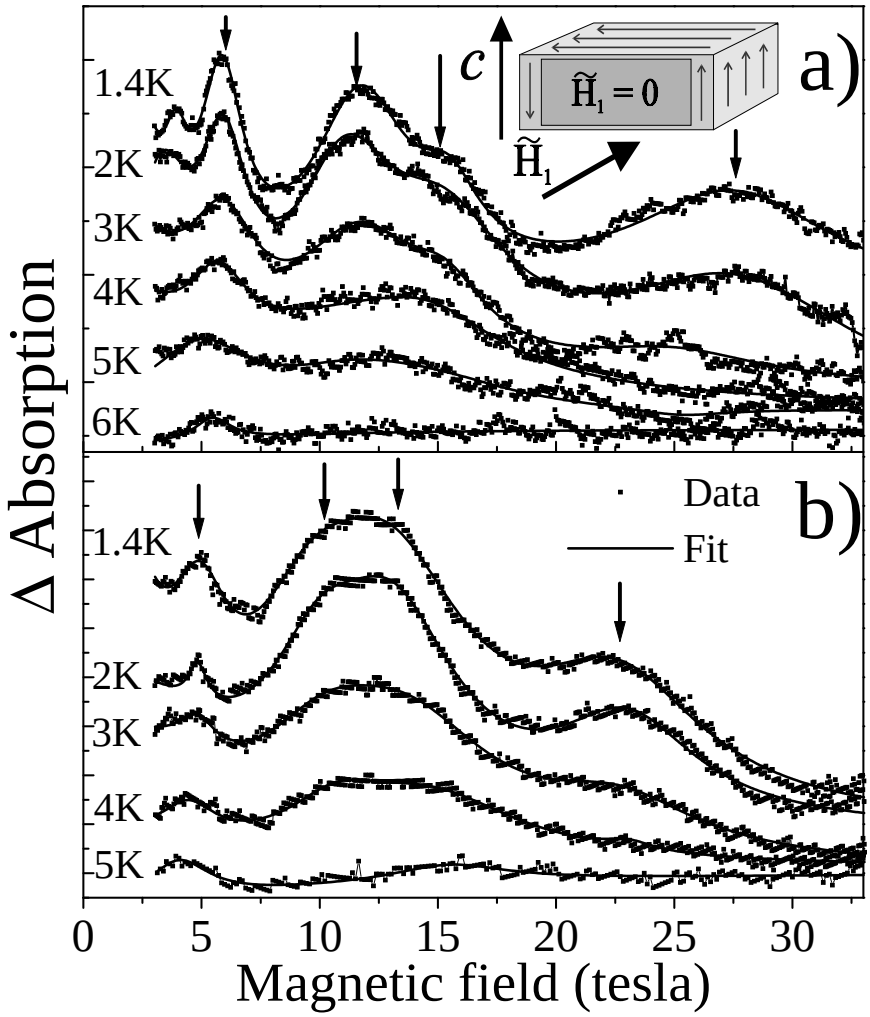

FIG. 1. Temperature and magnetic field dependence of the changes in absorption within the cavity (after a background subtraction) at a) 76.4 GHz, and b) 64 GHz; the data have been offset for the sake of clarity. See text for an explanation of the fitting procedure. Inset depicts the predicted AC currents at the sample surface

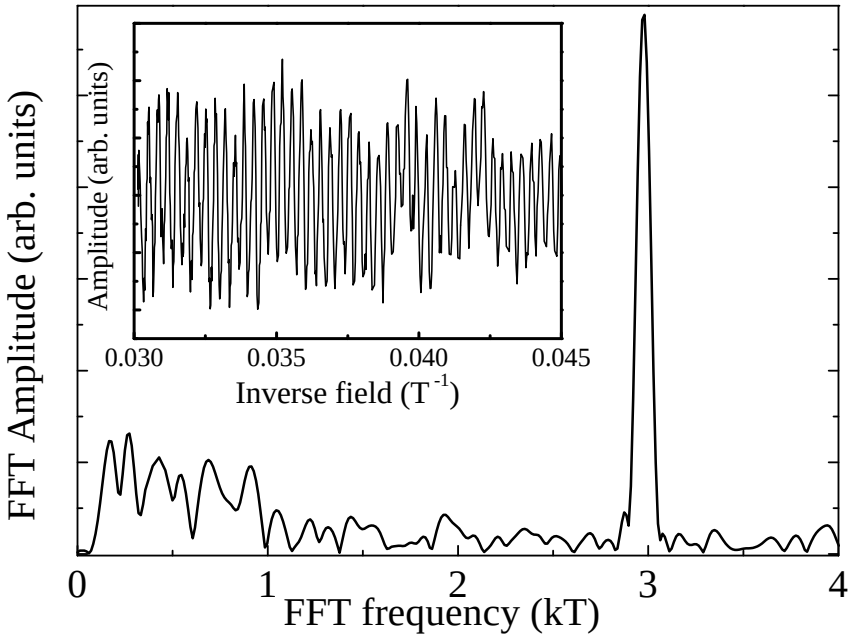

FIG. 2. Fast fourier transform (FFT) of the high-field cavity response (inset) at 44.5 GHz and at 1.4 K. SdH oscillations are visible in the inset which are periodic in $1/\mathbf{B}$. These oscillations correspond to the α-frequency observed in ref. [3].

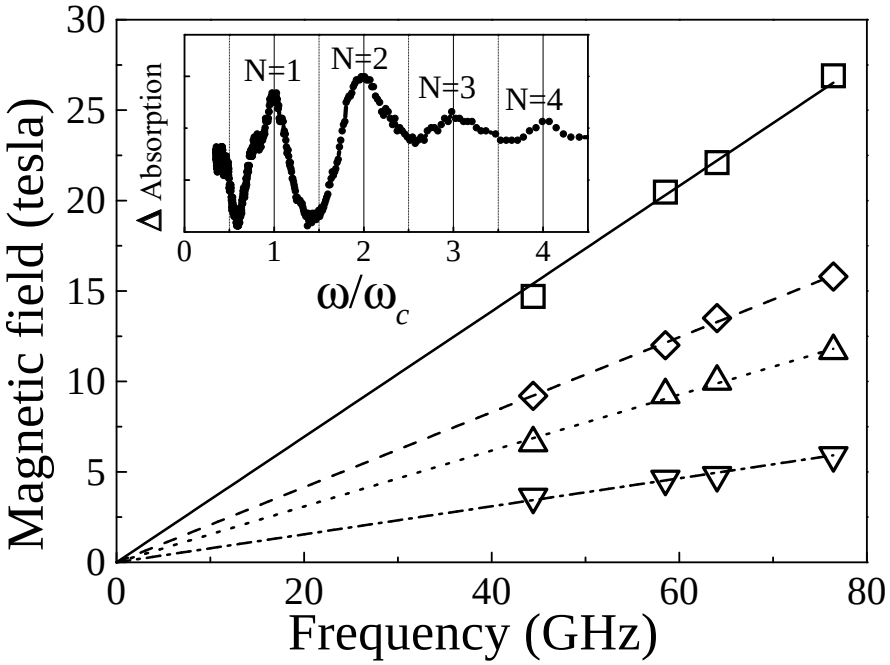

FIG. 3. Plots of the CR peak-field positions versus frequency at 1.4 K. We assign the resonances as follows: $\Box - \gamma$, $\diamondsuit - \beta$, $\triangle - \alpha$ 1st harmonic, and $\nabla - \alpha$ 2nd harmonic. The inset shows the α-series of CR harmonics at 76.4 GHz, plotted versus inverse field.