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Observation of quantum oscillations in the electrical resistivity of $SrRuO_3$

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We report the observation of quantum oscillations in the electrical resistivity of a high-quality thin film of the itinerant ferromagnet $SrRuO_3$. Our study demonstrates the existence of long-lived fermion quasiparticles at low temperatures, and strongly suggests that the ground state of $SrRuO_3$ is a Fermi liquid, even though ac and dc conductivity measurements at higher temperatures show anomalous metallic behavior. The implications of these results are discussed. [S0163-1829(98)52344-2]

Strongly correlated narrow-band oxides have been the subject of intense study in recent years. Diverse phenomena such as high-temperature superconductivity, metallic and insulating magnetism, and colossal magnetoresistance arise in these materials, often accompanied by unusual high-temperature metallic states, in which broad spectral features are observed in frequency-dependent properties such as optical conductivity and photoemission. Another commonly observed feature is ''bad metallic'' transport, whose signature is the existence of high-temperature conductivities that are so low that conventional analysis in terms of quasiparticles cannot be made.[1] In many of the materials, there is a strong suspicion that the metallic state cannot be described in terms of Fermi-liquid theory, but the formation of nonmetallic low-temperature states restricts the number of compounds in which this issue can be investigated in detail. Another complicating feature is the necessity to introduce fairly high levels of disorder to many oxide metals in order to introduce the charge carriers. For these reasons, narrow-band oxides containing low levels of disorder whose metallic state can be studied at low temperature are of great interest. Establishing the nature of the ground state in these materials will aid understanding of how the strange high-temperature states evolve, and should also give new perspectives about which, if any, of the metallic properties of materials such as the high-temperature superconductors signal the breakdown of Fermi-liquid theory.

Ruthenate metals give an ideal opportunity to address this issue. The subject of the present study, $SrRuO_3$, is a metallic ferromagnet ($T_c \sim 150$ K for thin films) with an orthorhombically distorted cubic perovskite structure.[2] Its saturation magnetic moment is thought to be approximately 1.6 μ_B, the largest of any $4d$ ferromagnet.[3] Mass enhancements indicated by specific heat studies reveal strong electronic correlations,[3,4] and critical behavior of the resistivity ρ near the Curie temperature has been interpreted as evidence for strong magnetic scattering.[5,6] Recent infrared conductivity measurements have found unusual frequency dependences to temperatures as low as 40 K,[7] while the room-temperature resistivity is high (230 $\mu\Omega$ cm), and continues to rise steadily to 1000 K.[4] This combination of observations has led to doubts about whether the metallic state can be described in terms of Fermi-liquid theory.[7]

A closely related ruthenate metal, the layered perovskite Sr_2RuO_4, has been studied extensively in recent years. Production of high-quality single crystals has revealed a disorder-sensitive superconducting state below 1.5 K,[8,9] which can be suppressed by modest magnetic fields. Quantum oscillation studies at high fields and low temperatures prove the existence of fermion quasiparticles,[10] and measurements of a number of bulk transport and thermodynamic properties give a consistent picture of a strongly anisotropic Fermi-liquid low-temperature metallic state,[11] in spite of bad metallic behavior at high temperatures.[12]

The question of whether $SrRuO_3$ is also a Fermi liquid motivated us to search for quantum oscillations in its electrical resistivity (the Shubnikhov–de Haas effect). The quantum oscillatory signal results from Landau quantization of the quasiparticles' cyclotron orbits in applied magnetic fields, but it is unobservable in most materials as it is heavily damped by impurity scattering. Although difficult, observation and analysis of the Shubnikhov–de Haas effect is a powerful way to establish the existence of long-lived fermion quasiparticles, because Fermi-liquid theory makes clear predictions about the field and temperature dependences of the oscillations.[13] We report the observation of oscillation frequencies of up to 3.5 kilotesla showing the Fermi-liquid temperature dependence, giving strong evidence that conven-

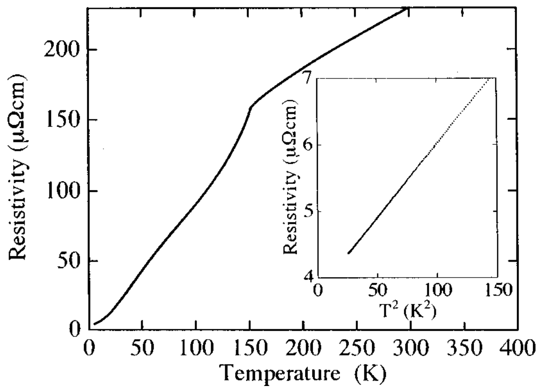

FIG. 1. The resistivity of the high-quality $SrRuO_3$ film used in this study. The inset shows that there is a T^2 scattering rate below approximately 10 K.

tional quasiparticles do indeed form at low temperatures in this three-dimensional oxide metal.

High-quality $SrRuO_3$ films were grown using an electron beam coevaporation technique on 2° miscut $SrTiO_3$ substrates described previously.[6] In general, resistance ratios of 35 are achieved; the film used in this study was the best grown to date, with a ratio of 60 between the resistivity at 300 K and that as $T \rightarrow 0$. Resistance measurements were performed using standard ac techniques with the specimen mounted on a dilution refrigerator (Oxford Instruments Kelvinox 25). Thermal contact to the mixing chamber was ensured by thermal anchoring of the leads to the sample, and excessive heating at the current contacts was avoided by using relatively low currents that generated power of less than 10 nW at the sample. The films are orthorhombic ($a = 5.53$ Å, $b = 5.57$ Å, $c = 7.85$ Å), with the (110) direction perpendicular to the surface. In these experiments, magnetic fields of up to 16 tesla were applied. Rotations of ±30° about the $(1\bar{1}0)$ direction were performed, with current always flowing along $(1\bar{1}0)$. Steps of 0.85° were used up to 9°, and then increased to 1.7° for higher angles. Unless otherwise stated, all data shown are for the orientation with the field along (110).

In Fig. 1 we show the resistivity of the as-grown film, measured in zero magnetic field. The ferromagnetic transition at 150 K results in a pronounced feature in the resistivity, as reported previously.[14] Below approximately 10 K, the T^2 scattering rate expected in a Fermi liquid is observed (inset). Figure 2 shows the basic features of the low-temperature magnetoresistance, measured at 35 mK as the field is swept from 14 to -14 tesla. The sharp peak at approximately -0.2 tesla is a reproducible hysteretic feature associated with the coercive field in this ferromagnetic material. Reversing the direction of the field sweep reverses the sign of the field at which it is observed (not shown). The other features on the curve are reversible and symmetric in field. There is a point of inflection at ~2 tesla, and another one at ~5 tesla followed at higher fields by a quasilinear field dependence with some structure. Subtracting a smooth background from the data between 5 and 14 tesla gives the oscillatory signal shown in the inset to Fig. 2.[15] The main component is periodic in reciprocal field, and has a very low frequency of 34 tesla.

The central findings of our study are summarized in Figs. 3 and 4. High precision data taken during slow field sweeps between 11 and 16 tesla show the existence of quantum oscillations with much higher frequencies; an example is shown in Fig. 3. As seen in Fig. 4, two frequency components at 1.5 and 3.5 kilotesla have been resolved. The oscillatory amplitude is extremely small, corresponding to a resistivity of less than 1 nΩ cm or approximately 1 part in 10^4 of the total resistivity shown in Fig. 2. The frequencies of both orbits have very weak angular dependence; only a few percent over the angles for which they are observable. The 1.5 kilotesla orbit has a strong angular dependence of its amplitude, and can be resolved only within ±3° of (110), while the angular dependence of the amplitude of the 3.5 kilo tesla orbit is much weaker.[16]

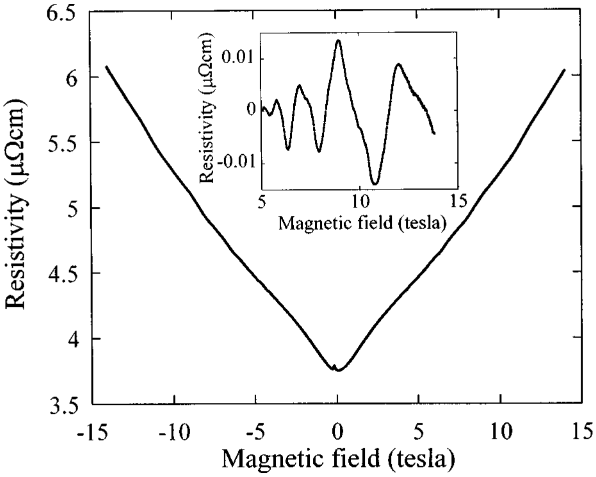

FIG. 2. The magnetoresistance of $SrRuO_3$ measured as the magnetic field is swept from 14 to -14 tesla, at a temperature of 35 mK. The peak at -0.2 tesla is a hysteretic feature associated with the coercive field of this itinerant ferromagnet. The inset shows oscillatory behavior between 5 and 15 tesla. The oscillations are periodic in reciprocal field, with a frequency of 34 tesla.

The temperature dependence of the amplitudes of each frequency component are shown in the insets to Fig. 4. In a noninteracting system, the electron occupation is the well-known Fermi function. In this case, the temperature dependence of quantum oscillations should have the functional form of the Fourier transform of the energy derivative of the

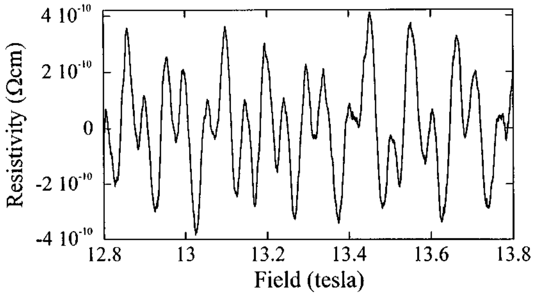

FIG. 3. High-frequency Shubnikov–de Haas oscillations of very small amplitude (approximately 1 part in 10^4 of the total resistivity) can be observed at high fields. To remove the strongly field-dependent background seen in Fig. 2, a filtering procedure that strongly attenuates any frequencies below 750 tesla has been used. The measurement temperature was 35 mK.

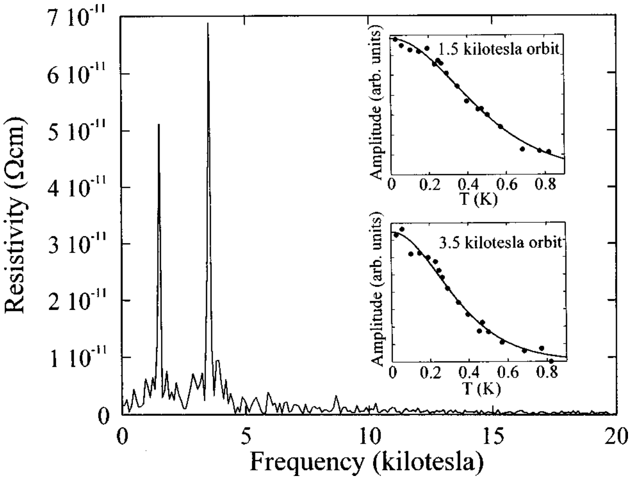

FIG. 4. The square root of the power spectrum of the data shown in Fig. 3. Two main peaks are clearly resolved, at 1.5- and 3.5 kilotesla, respectively. The insets show the temperature dependences of the two main components. The standard Fermi-liquid predictions (solid lines) fit the data well, confirming the existence of conventional fermion quasiparticles.

Fermi function at temperature T, i.e., $X/\sinh X$, where $X=2\pi^2 k_B T/\hbar\omega_c$, $\omega_c=eB/m^*$ is the cyclotron frequency and m^* is the cyclotron effective mass.[17] The same holds for a Fermi liquid because the quasiparticle occupation also follows the Fermi distribution at low temperature, and the quasiparticles decay sufficiently slowly (quadratically in energy away from the Fermi surface.[18]) As can be seen in the insets to Fig. 4, the data agree with the Fermi-liquid prediction of the temperature dependence to within experimental accuracy, giving strong evidence for the existence of long-lived fermion quasiparticles in $SrRuO_3$ at low temperatures. The measured cyclotron masses are 4.5 and 6.1 electron masses for the 1.5 and 3.5 kilotesla orbits, respectively.

From the above observations, it is reasonable to conclude that the low-temperature metallic state of $SrRuO_3$ can be understood in terms of Fermi-liquid theory, in spite of good evidence for unusual metallic behavior at temperatures of 40 K and higher. The present work suggests that the crossover between the two regimes occurs at approximately 10 K (inset to Fig. 1), and $SrRuO_3$ thus gives an ideal opportunity to investigate the relationship between the two regimes in a three-dimensional material.

The details of the observations present further interesting challenges to our understanding. Electronic band-structure calculations for $SrRuO_3$ have been performed by several groups,[4,19–21] including a full potential local spin density calculation by Singh.[19] The Fermi surface is predicted to be very complicated, with up to nine bands (three majority spin and six minority spin) crossing the Fermi level, in contrast to the two main orbits reported here. In quantum oscillation experiments, the signal from large orbits is damped more strongly by impurity scattering than that from smaller orbits, so it is not unusual for large orbits to be unobserved. In $SrRuO_3$, however, the orthorhombic distortion leads to a small Brillouin zone, and the high-frequency orbits that we observe are comparable to the largest predicted by the band-structure calculations,[22] making it harder to account for the ‘‘missing orbits.’’ It seems that more experimental and theoretical work is desirable to establish the extent to which quantum oscillation measurements and band-structure calculations are in detailed agreement in $SrRuO_3$. It is important that attempts are made to do this, as precise knowledge of the low-temperature electronic structure is likely to be a necessary precursor to understanding the crossover to unusual high-temperature behavior.[23,24]

We now return to the much stronger signal that we have observed at a frequency of 34 tesla. If the basic oscillation (whose frequency is again only weakly dependent on angle) is analyzed as a standard Fermi surface pocket, it has a tiny volume (less than 0.1% of that of the largest pocket). By 15 tesla, it is quantized into fewer than five Landau levels. The signal is weakly temperature dependent; measurements at 35 mK, 4.2 K, and 10 K yield an estimated cyclotron mass of 0.2 electron masses. It is fairly unusual for such a small pocket to coexist with large Fermi surface pockets, and the observation is worthy of further theoretical attention. For example, the additional structure seen at fields above 10 tesla is not accounted for in the simple interpretation given here.

In summary, we have observed quantum oscillations in the resistivity of the itinerant ferromagnet $SrRuO_3$. The highest-frequency oscillations correspond to extremal Fermi surface areas that are comparable with the largest predicted by electronic energy band-structure calculations, and their temperature dependences follow the standard Fermi-liquid form.[24] These observations, combined with a quadratic temperature dependence of the electrical resistivity below 10 K, lead us to conclude that the low-temperature behavior of $SrRuO_3$ can be understood in terms of Fermi-liquid theory, in spite of the range of bad metallic properties that are seen at medium and high temperatures. It is thus an ideal material in which to study the temperature-dependent crossover between the two regimes in three dimensions. In addition, several details of our data motivate further theoretical and experimental investigation.

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[14] The Curie temperature of the thin films is about 10 K lower than that of single crystals (Refs. 3 and 4), presumably because of strains introduced by the miscut substrate.

[15] A second-order polynomial was used to model the background.

[16] The thin-film form of the sample was crucial to our being able to detect these tiny resistive oscillations, but it constrains the possibility of a full angular study, because the finite thickness (~1000 Å) means that for the 3.5-kilotesla signal, the sample dimension in the plane of the quasiparticle orbit becomes comparable to the orbit diameter as the angle is increased, consistent with observed amplitude attenuation at field angles beyond 20° from (110). If it proves to be possible to observe de Haas–van Alphen oscillations in single crystals, this difficulty could be overcome.

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[23] It will be necessary to perform a full rotation study (see Ref. 16) and compare the results with a band-structure calculation that includes spin-orbit coupling and the dependence of that coupling on the angle between the field and the crystallographic axes, an effect that can be important in ferromagnets (Ref. 24). Our existing data give preliminary evidence for two additional frequencies at 300 and 700 tesla, which await confirmation. At some angles, slight splitting of the 3.5-kilotesla peak is also seen. None of these, however, seems to offer a simple resolution of the discrepancy with the existing calculations.

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