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Tunneling magnetoresistance and quantum oscillations in bilayered $Ca_3Ru_2O_7$

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We report the interplane resistivity, ρ_c, at high magnetic fields B, with different orientations together with structural and magnetic properties of bilayered $Ca_3Ru_2O_7$, a Mott-like system with a gap of 0.1 eV. A wide array of conventionally unanticipated phenomena revealed in this work includes (1) a collapse of the c-axis lattice parameter at a metal-nonmetal transition, (2) quantum oscillations in ρ_c in the gapped, nonmetallic state for the $B\|c$ axis, (3) interplane tunneling magnetoresistivity for the $B\|a$ or b axis, and yet conspicuously different anisotropies of the colossal magnetoresistivity and magnetization, and (4) a non-Fermi-liquid behavior in a metallic state fully recovered in high magnetic fields. The implications of the coexistence of these conflicting phenomena are discussed.

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Materials that are characterized by a strong interplay between different degrees of freedom tend to exhibit physical phenomena that are astonishing, exotic, yet cannot be understood with conventional notions. This viewpoint has been spectacularly demonstrated by the high-temperature superconducting copper oxides, and later, the colossal magnetoresistance (CMR) manganese oxides. Studies of these and other $3d$-electron-based transition metal oxides have uncovered a wealth of fascinating physics that has challenged our fundamental understanding of materials that cannot be described by the Landau-Fermi liquid theory, a frequently used model for metals where Coulomb repulsion between electrons is incorporated into effective masses and couplings. Ruthenium oxides or ruthenates as a new class of materials with characteristics of highly correlated electrons have increasingly captured attention in recent years,[1] but the $4d$-electron-based ruthenates are still by and large an uncharted territory rich with interesting physical properties that very often deviate from those of other materials including even $3d$-electron transition-metal oxides. Here the exotic phenomena observed in the bilayered $Ca_3Ru_2O_7$ provide another striking example that defies conventional wisdom.

$Ca_3Ru_2O_7$ is a layered system[2,3] with a charge gap of 0.1 eV (Ref. 4) that bears a resemblance to a Mott system.[2–7] It is known that $Ca_3Ru_2O_7$ undergoes an antiferromagnetic ordering at $T_N = 56$ K followed by a first-order metal-nonmetal transition at $T_{MI} = 48$ K.[2] $Ca_3Ru_2O_7$ also features first-order metamagnetic and magnetoresistive transitions that lead to a field induced ferromagnetic (FM) metallic phase below T_{MI} where spins are almost fully polarized along the a axis, the magnetic easy-axis, at $B = 6$ T.[2,3] In addition, the electron scattering rate is strong;[5] thus, there is a remarkably short mean free path l at room temperature which is highly anisotropic, ranging from 0.8 to 8 Å, well beyond the limit for bandlike transport, assuming a typical Fermi velocity of $10^7 - 10^8$ cm/s.

In this paper we describe and discuss the c axis or interplane resistivity ρ_c at high magnetic fields with different orientations together with magnetization and the c-axis lattice parameter as a function of temperature in $Ca_3Ru_2O_7$.[8] All results underscore a critical role of unusually strong coupling between spin, charge, t_{2g} orbital and lattice degrees of freedom that drives a wealth of physical phenomena, which are conventionally unexpected.

Shown in Fig. 1 are the interplane resistivity ρ_c and the lattice parameter for the c axis (right scale) as a function of temperature T. The data obtained from x-ray diffraction exhibits a rapid decrease in the c-axis lattice parameter at T_{MI},

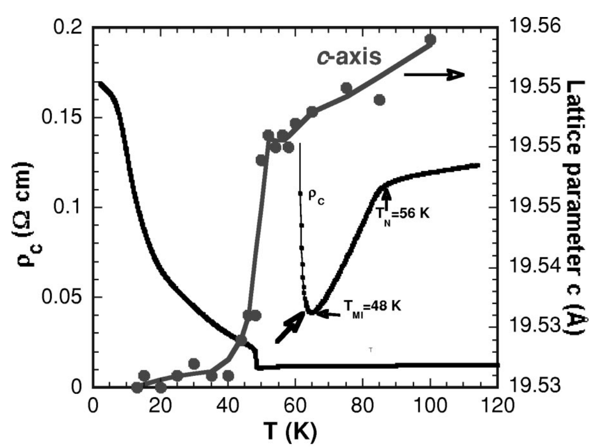

FIG. 1. The interplane resistivity ρ_c (left scale) and the c axis vs temperature T; the inset is the enlarged ρ_c near T_M.

but no systematic changes in the *ab* plane. The simultaneous structural, electronic and magnetic[2,3] transitions at $T_{\rm MI}$ unambiguously indicate a strong charge-spin-lattice coupling, which is also evidenced in the Raman study, where $T_{\rm MI}$ is found to be associated with an abrupt softening of a *c*-axis phonon mode[4] sensitive to pressure.[7] It appears quite surprising that the collapse of the *c*-axis lattice parameter, which would be expected to enhance the overlap of orbitals, does not lead to a more metallic state, but conversely, a gapped, nonmetallic ground state, as evidenced by an abrupt metal-nonmetal transition at $T_{\rm MI}=48$ K followed by a rapid increase in ρ_c by a factor of 18 [$T_{\rm MI}$ occurs in ρ_a and ρ_b as well, which change by about a factor of 5 (Refs. 2 and 3)]. It is possible that the collapse of the *c*-axis lattice parameter may be associated with a Jahn-Teller-like distortion of the Ru-O octahedra, which lifts the degeneracy of the t_{2g} orbitals by lowering the energy of the d_{xy} orbital relative to that of the d_{yz} and d_{xz} orbitals. Recent first-principles calculations for Sr_2RuO_4 indicate that the shortening of the *c* axis or the flattening of RuO_6, which results in orbital polarization and reducing bands of three t_{2g} states, is the key fact in stabilizing the insulating magnetic ground state.[9] This point may also be valid for $Ca_3Ru_2O_7$, as both $T_{\rm MI}$ and magnitude of ρ_c are found to rise drastically even at low uniaxial pressure applied along the *c*-axis in our recent study. For instance, at 2.5 kbar, $T_{\rm MI}$ increases to 73 K and ρ_c jumps by more than two orders of magnitude.[10] In contrast, the uniaxial stress along the *a* axis and hydrostatic pressure leads to a decrease in $T_{\rm MI}$ and resistivity,[11] consistent with the recent Raman results.[7]

Given such a nonmetallic state, the quantum oscillations (QOs) in ρ_c is unexpectedly observed for $20\,\mathrm{mK}<T<6.5$ K when $B\|c$, as illustrated in Fig. 2(a) where ρ_c vs B is plotted. The amplitude of the QOs as a function of inverse field B^{-1} is presented in Fig. 2(b) for several temperatures. The inset shows the amplitude of the oscillation signal normalized by T in a logarithmic scale as a function of T. The solid line is a fit to the Lifshitz-Kosevich formulas, $x\sinh x$, where $x=14.69\ \mu_c T/B$. The fit yields a cyclotronic effective mass $\mu_c=0.85\pm0.05$, seemingly suggesting the existence of fermion quasiparticles.[12] Markedly, this cyclotronic effective mass is different from the enhanced thermodynamic effective mass estimated from the electronic contribution γ, to the specific heat.[2] This apparent disagreement between the thermodynamic effective mass and the cyclotronic effective mass is quite common in heavy fermion systems,[13] and is attributed to the fact that it has been difficult to resolve the higher effective masses in quantum oscillation experiments. It is possible that some portion of the Fermi surface, if any, with larger masses might have not been detected in our measurements. Conspicuously, the QOs are absent in resistivity for the *a* and *b* axes when $B\|c$. Analyses[14] reveal an extremely low frequency of $f_1=28$ T, which, based on crystallographic data[3] and the Onsager relation $F_0=A(h/4\pi^2 e)$ (*e* is the electron charge), would correspond to an area of only 0.2% of the first Brillouin zone, as shown in Fig. 2(b) where the thin lines outline beating between two frequencies ($f_2=28$ T and $f_2=10$ T). In addition, the disappearance of the QOs at $B_{\|c}>25$ T may indicate the proximity of the quantum limit,[14]

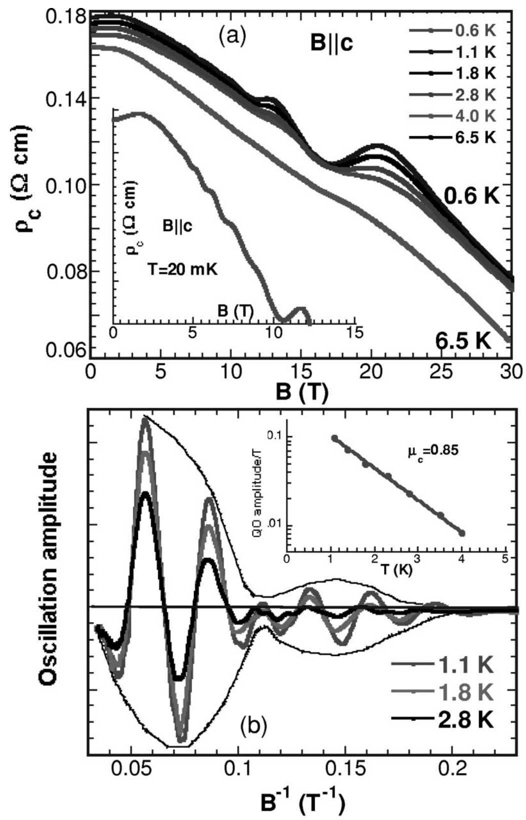

FIG. 2. (a) ρ_c vs $B\|c$ axis for various *T*, as indicated; the inset is ρ_c vs $B\|c$ axis for $T=20$ mK at lower *B*. Note the development of the QOs with decreasing *T* (the QO signal is defined as $(\sigma-\sigma_b)/\sigma_b$ where σ is the conductivity (or the inverse of ρ_c) and σ_b is the background conductivity). (b) The amplitude of the oscillations vs B^{-1} for different *T*, the thin lines are a guide to the eye to show beating between f_1 and f_2; the inset is the QO amplitude normalized by *T* vs *T*. The solid line is a fit to the LK formulas.

which imposes limitations on the applicability of the Lifshitz-Kosevich formalism[15,16] to the data presented here. However, when *B* is larger than the first-order metamagnetic transition (6 T) and within the *ac*-plane (not parallel to either the *a* or *c* axis), the QOs reoccur, showing a larger frequency of 47 T. In contrast, no oscillations are discerned for *B* in the *bc* plane even though ρ_c in this configuration is smaller by as much as two orders of magnitude than that for *B* in the *ac* plane. The difference suggests a critical role of the first-order metamagnetic transitions, which could alter the Fermi surface in favor of the reoccurrence of the QOs.[10]

Quantum oscillations are a trademark of a Fermi liquid. The absence of QOs in the vast majority of transition-metal oxides has been taken as an additional evidence of non-Fermi liquids. The occurrence of the QOs in a gapped, nonmetallic oxide such as $Ca_3Ru_2O_7$ is therefore intriguing in that conventional physics requires the existence of a Fermi surface (FS) (a metallic state) and a long mean free path ($>10^3$ Å) for QOs to occur. In this case it may well be the manifestation of new physics associated with the multiorbital nature of the electronic properties.[17,18] It has been proposed that a situation may occur in which electrons in the d_{xz} and d_{yz} orbitals undergo a Mott transition while those in the d_{xy}

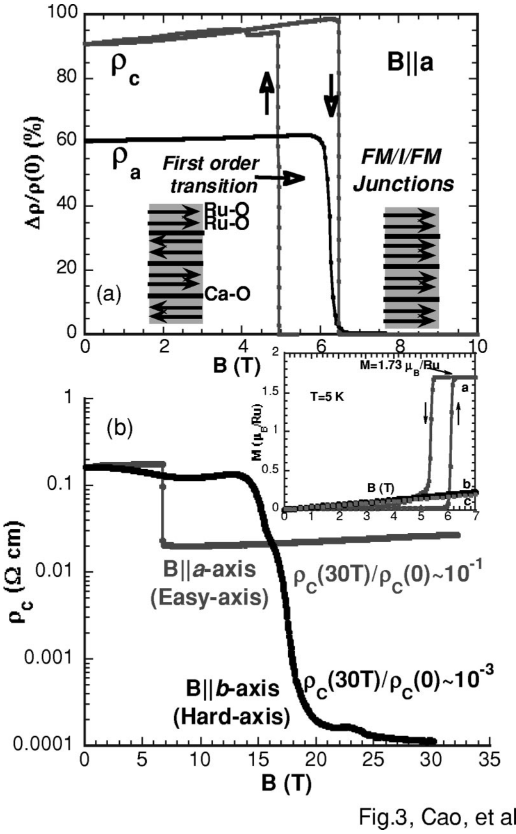

FIG. 3. (a) The magnetoresistivity ratio $\Delta\rho/\rho(0)$ $[\Delta\rho=\rho(0)-\rho(B)]$ vs $B\|a$ axis for both ρ_c and the a axis ρ_a at $T=0.6$ K. Note that at $B>6$ T the system is virtual FM/I/FM junctions. (b) $\rho_c(B)$ in log scale for B parallel to the easy axis (a axis) and B parallel to the hard axis (b axis). Note that the drop in ρ_c for B parallel to the easy axis is two orders of magnitude *smaller* than, that for B parallel to the hard axis, i.e., $\rho_c(30\ \mathrm{T})/\rho_c(0)\sim10^{-1}$ for $B\|a$ but 10^{-3} for $B\|b$ at $T=0.6$ K. The inset shows the anisotropy of M contradictory to that of $\rho_c(B)$.

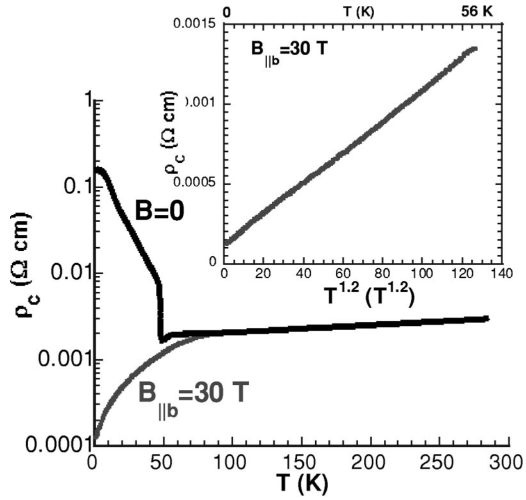

FIG. 4. ρ_c in a log scale at $B=0$ and 30 T as a function of T for the $B\|b$ axis. The inset shows ρ_c at $B_{\|b}=30$ T as a function of $T^{1.2}$.

orbitals do not.[18] Band structure calculations on $Sr_3Ru_2O_7$ also show that a tetragonal-orthorhombic phase transition naturally leads to small lens-shaped Fermi surface pockets that originate from the d_{xy} orbitals.[19] Therefore, one of possible scenarios could be that extremely small FS pockets form with a low carrier density and, thus, a long mean free path,[20] therefore it might be possible for the QOs to occur. However, it is puzzling that no QOs have been discerned in resistivity for the a and b axes when $B\|c$, and, more generally, in other systems with a low density of states. It is noted that QOs have been observed in one-dimensional density-wave-like systems,[21–23] but, given the higher dimensionality, the analogy may not be entirely valid in $Ca_3Ru_2O_7$. It is therefore not entirely clear whether or not the QOs are a Fermi-surface phenomenon. Another possible scenario could be that the observed QOs are associated with a tunneling effect due to the layered nature of the system. Apparently, the QOs manifest an unusual phenomenon that merits more attention and further investigations.

The QOs are immediately overpowered by a precipitous drop in ρ_c as B rotates away from the c axis. Shown in Fig. 3(a) is the magnetoresistivity ratio defined as $\Delta\rho/\rho(0)[\Delta\rho=\rho(0)-\rho(B)]$ as a function of $B\|a$ axis for both ρ_c and the a axis ρ_a at $T=0.6$ K. As seen, the abrupt metamagnetic transition at 6 T leads to a $\Delta\rho_c/\rho_c(0)$ of more than 90% at $B>6$ T for ρ_c, much larger than $\Delta\rho_a/\rho_a(0)$ of 60% for ρ_a. The larger interplane CMR is believed to be due to a tunneling effect facilitated by a field-induced coherent motion of spin-polarized electrons between Ru-O planes. Because of the layered nature, the spin-polarized Ru-O planes sandwiched between insulating (I) Ca-O planes form an array of FM/I/FM junctions [see Fig. 3(a)] that enhances the probability of tunneling and thus electronic conductivity. It is noted that the current-perpendicular-to-plane giant magnetoresistance (GMR) could be larger than the current-in-plane GMR due to spin filtering through nonmagnetic layers.[24]

However, what is entirely unexpected is that, as shown in Fig. 3(b), the drop in ρ_c for B parallel to the easy axis (a axis) is two orders of magnitude *smaller* than that for B parallel to the hard axis (b axis), i.e., $\rho_c(30\ \mathrm{T})/\rho_c(0)\sim10^{-1}$ for $B\|a$ but 10^{-3} for $B\|b$ at $T=0.6$ K. This is completely contrary to the anisotropy of the magnetization (see the inset). The negative magnetoresistance is in general driven by the reduction of spin scattering.[25] The different in-plane anisotropies for the magnetization and the CMR clearly suggest that spin-polarization alone cannot at all account for the CMR in this system. The spin-orbit interaction, which mixes states, could lead to an anisotropic scattering, and, thus, an anisotropic magnetoresistance. However, the effects of the spin-orbit interaction are small in general, and are thus unlikely to be responsible for the difference of two orders of magnitude in the resistivity. Given the unusually strong magnetoelastic effects so evidenced in the system,[10] the lattice and orbital degrees of freedom may play a critical role in the scattering mechanism(s). Such an unusual scattering behavior certainly merits more experimental and theoretical investigations.

It also comes as a surprise that even though ρ_c drops by three orders of magnitude as displayed in Fig. 4, and shows a fully metallic state when $B\|b$, the temperature dependence of ρ_c at $B_{\parallel b}=30$ T does not at all obey the T^2 dependence expected for a Fermi liquid. Instead, ρ_c exhibits an unusual $T^{1.2}$ dependence (see the inset) in the fully spin-polarized state where the spin degree of freedom should be eliminated. The low power-law temperature dependence of ρ normally implies strong scattering. While magnon scattering might partially account for this behavior, the anomalous temperature dependence of ρ_c once again suggests an unusual scattering mechanism(s) that governs not only the ground state but also persists well into the high temperature regime.

The results presented here illustrate the critical role of the collapse of the *c*-axis lattice parameter for the presence of the antiferromagnetic nonmetallic ground state where the different kinds of ordering, which are conventionally expected to exclude each other, seem to be characteristically synergistic in $Ca_3Ru_2O_7$. The extraordinary coexistence of both nonmetallic and fermion-quasiparticle characteristics, which was recently found to be hardly uncommon in the layered ruthenates,[26] calls for innovative theoretical approaches to the extended, correlated electrons; the drastically different anisotropies of the CMR and the magnetization, together with the non-Fermi liquid of ρ_c in the field-induced metallic state, signals an unusual scattering mechanism other than the spin-scattering-dominated mechanism that works marvelously well in other materials. To address these profound problems will surely broaden and deepen our understanding of fundamentals of correlated electrons.

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[1] See, for instance, Y. Maeno, T. M. Rice and M. Sigrist, Phys. Today **54** (1), 42 (2001).
[2] G. Cao *et al.*, Phys. Rev. Lett. **78**, 1751 (1997).
[3] G. Cao *et al.*, Phys. Rev. B **62**, 998 (2000).
[4] H. L. Liu *et al.*, Phys. Rev. B **60**, R6980 (1999).
[5] A. V. Puchkov *et al.*, Phys. Rev. Lett. **81**, 2747 (1998).
[6] R. P. Guertin *et al.*, Solid State Commun. **107**, 263 (1998).
[7] C. S. Snow, S. L. Cooper, G. Cao, J. E. Crow, S. Nakatsuji, and Y. Maeno, Phys. Rev. Lett. **89**, 226401 (2002).
[8] The single crystals of $Ca_3Ru_2O_7$ were grown using the flux methods [Refs. 2 and 3] and characterized by x-ray diffraction, SEM, and TEM. The crystals studied are of high quality with a Dingle temperature (a measure of defects, disorder and impurities), T_D, ranging from 1.5, and 3.0 K, comparable to those of high-quality organic metals whose T_D varies from 0.5 to 3.5 K.
[9] Z. Fang and K. Terakura, Phys. Rev. B **64**, 020509(R) (2001).
[10] G. Cao *et al.* (unpublished).
[11] The results of the comprehensive pressure study will be published elsewhere.
[12] G. Cao *et al.*, Phys. Rev. B **56**, 5387 (1997).
[13] N. Harrison *et al.*, Phys. Rev. B **63**, 081101(R) (2001).
[14] The usual fast Fourier analysis becomes unreliable because of the existence of only a few oscillations. The frequencies can, however, be determined by directly measuring the period of the oscillations. The quantum limit is expected when the quantized magnetic energy $\mathcal{E}=(n+1/2)heB/2\pi mc$ becomes comparable to the electronic or Fermi energy.
[15] I. M. Lifshitz and A. M. Kosevich, Zh. Eksp. Teor. Fiz. **29**, 730 (1956) [Sov. Phys. JETP **2**, 636 (1956)].
[16] D. Schoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, 1984).
[17] T. Hotta and E. Dagotto, Phys. Rev. Lett. **88**, 017201 (2001).
[18] V. I. Anisimov *et al.*, Eur. Phys. J. B **25**, 191 (2002).
[19] D. J. Singh and I. I. Mazin, Phys. Rev. B **63**, 165101 (2001).
[20] From the frequency estimated, one can calculate the area of the orbit through the Onsager relation $F=(h/4\pi^2e)$, which provides an estimate for the Fermi wave vector, $A/\pi\cong k_F^2$ leading to $k_F\cong 1.5\times 10^8$ m^{-1}, a quite small value. From this value we can estimate the mean free path $l=\nu_F\tau$, where ν_F is the Fermi velocity, $\nu_F=hk_F/2\pi m$, and τ is obtained through the residual resistivity $\tau=\mathrm{m}/ne^2\rho$ at low T. These expressions allow an estimate of the Mott-Ioffe-Regel limit for coherent transport in a given system, $k_F l=3\pi h/2e^2k_F\rho$, which depends only on the residual resistivity and on k_F, and gives rise to $k_F l\gg 1$, where l is large and k_F rather small for the gapped state, in agreement with the large values for τ obtained from the Dingle factor.
[21] S. Hill *et al.*, Phys. Rev. B **58**, 10 778 (1998).
[22] W. Kang *et al.*, Phys. Rev. Lett. **70**, 3091 (1993).
[23] N. Biskup *et al.*, Phys. Rev. B **50**, 12 721 (1994).
[24] S. Sanvito *et al.*, Phys. Rev. B **60**, 7385 (1999).
[25] Y. Tokura, *Colossal Magnetoresistive Oxides* (Gordon and Breach, New York, 2000).
[26] We have recently observed the QOs in other non-Fermi-liquid ruthenates such as $BaRuO_3$ and $Sr_4Ru_3O_{10}$. The results are to be published elsewhere.